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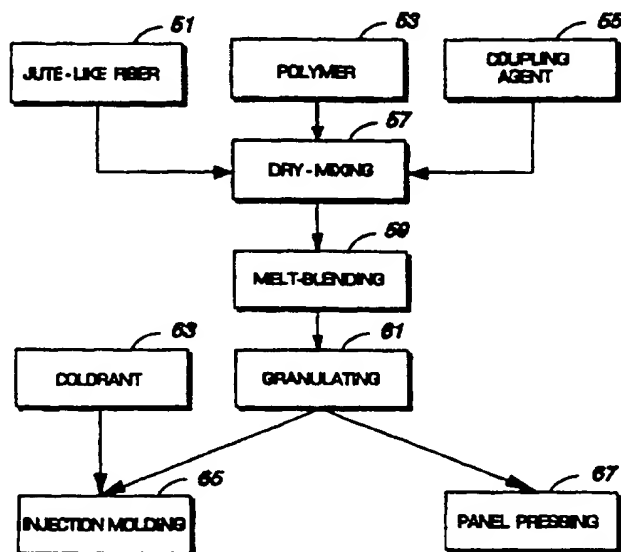
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(54) Title: JUTE AND KENAF FIBER COMPOSITE MATERIALS AND METHODS FOR PRODUCING SAME



## (57) Abstract

Fiber-reinforced structures comprising a thermoplastic polymer (53) and containing between 30 % and 60 % by weight of reinforcing filaments arrayed randomly or aligned within the structure have been produced in a continuous process and have high cantilever, tensile and flexural strength, high stiffness, low creep and low relative cost. The high strength and stiffness result from compatibilizing of natural fibers (51) and the thermoplastic matrix (53) by wetting or coating of fibers (51) between 1 mm and 20 cm in length with maleated polypropylene coupling agents (55) and, optionally, with a small volume of phenolic or epoxy resins. The mixed material is further processed by molding (65) under conditions in which material temperatures are maintained below 230 °C and pressure is held below 28,000 psi.

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## **JUTE AND KENAF FIBER COMPOSITE MATERIALS AND METHODS FOR PRODUCING SAME**

This case is a continuation-in-part of U.S. Patent Application 08/287,217 filed on August 8, 1994.

### **Field of the Invention**

The present invention relates to the field of fiber reinforced structures and more particularly to the use of jute and kenaf fibers to reinforce thermoplastic compositions including polypropylene.

### **Background of the Invention**

Manufacturing materials of many types are employed in various services for the production of structures and packaging. For example, containers are often made of wood for its strength and weight characteristics, but it has low tolerance to its environment where water, through absorption, and other agents may cause rotting and denigration of its integrity and strength. Thus, the structure's life cycle is shortened requiring further wood resources to make new containers and creating a disposal problem for the wood from previously used containers. Further, most wood product containers are hammered shut and pried open repeatedly, which tends to wear out the product and increase its susceptibility to rotting and breakdown.

Because of the shorter life of wood products, other advantages cannot economically be taken when employing the wood for use as a structural material, and especially where shaping the material is concerned. The structural shaping which would result in weight loss without significant increase in structural strength, is simply not practical, and would require more wood material to be consumed. Such

structural shaping would have to occur through precision CNC milling or other cutting techniques.

Agro-wastes and wood flour have played an important role in the plastics industry. The earliest phenolics were mixed with wood flour to reduce cost and improve the processing of the resin. The use of lignocellulosic material as fillers in thermoplastics such as polyethylene and polypropylene has, however, been limited, because of difficulties associated with surface instructions. The inherent polar and hydrophilic nature of the fibers and the nonpolar characteristics of the polyolefins result in difficulties in compounding of these materials and poor-stress-transfer efficiencies. Commonly used coupling agents in fiber-composite production do not function efficiently in lignocellulosic-polyolefin systems and may be too expensive

Plastics are increasingly employed in place of wood in many structures. Plastics offer the advantages of enhanced resistance to the ambient environment. Some plastics, however, can harden, crack, and suffer loss in performance and damage to structural characteristics. Certain plastics, known as high-end plastics, have higher resistance to aging and environmental effects as well as improved structural characteristics. Their extremely high cost, however, make them impractical for use in most applications. Application of high-end plastics is further limited by their high material density which can often result in unacceptably high product weight and heft.

Creep is a major issue with structural material manufactured of thermoplastics. Under load conditions, the popular low-end structural thermoplastics, primarily polypropylene and high density polyethylene, experience significant creep. This renders the unreinforced material unsuitable for a wide range of industrial and commercial applications - including stackable 55 gallon containers

and large high-load stackable boxes and pallets. Product creep is the result of creep in single constituents (fiber and/or matrix), bonding strength, rupture of fibers, matrix interface and degree of fiber overlap. The key issue with respect to creep is a finding the level at which creep is reversible. This is the level at which the transition to non-linear inelastic behavior occurs - usually a strain level of about 0.8%.

With plastics, the structural shapes which enhance the strength to weight ratio of products and material are obtained by molding. Molded materials have significant advantages over carved, cut, hewn or otherwise reduction-formed material in that there are no dead loss of material, such as shavings, sawdust, and odd pieces. This is a significant disadvantage of wood. This moldability advantage of plastics, however, is still insufficient to overcome the cost and/or inherent structural limitations of plastics in many applications. High-end plastics are still too expensive, and low end plastics do not, themselves, have adequate structural properties.

One proposed solution has been the addition of filler material within the volume of the plastics to add stiffness and/or strength. Many types of materials have been tried, including paper, wood flour, wood chips, glass fibers, mica, calcium carbonate, talc, and wollastanite, among others. Not all of the filler/reinforcer materials actually enhance the structural properties of the resulting composite. Those which do will typically add dramatically to cost.

Since plastics are non-polar, fillers such as wood and paper are difficult to bond to the plastic material. In such cases, the filler may actually cause the resulting composite to have inferior characteristics with respect to the use of the plastic alone. The principal advantage in the use of such materials becomes that of extending the more expensive plastic material with a less expensive filler and, perhaps, to create enhanced stiffness in the final molded product. In such instances the finished product

will typically have lower tensile strength characteristics than the underlying thermoplastic matrix.

Materials of high strength, such as carbon fibers, glass fiber, mica, Wollastanite (calcium metasilicate [ $\text{CaSiO}_3$ ]) and other material, bond more readily with thermoplastics to produce a high strength composite, but they add prohibitively to the cost. Such composites are fine for, high-valued specialty applications where material costs represent only a minor element in the total item cost (or value), or such applications, as in airplane and rocket parts, where item cost is relatively unimportant, and strength-for-weight is an overriding consideration. Use of these materials in large, high volume applications is typically prohibitive.

Some of the more popular fillers/reinforcers are also highly abrasive materials. Significant damage can be caused to compounding and molding equipment. The dramatically high price of metal molds and dies can make this type of damage extremely costly. Abrasiveness is also an inherent quality problem in the finished product. Once the smooth finish on the abraasive-reinforced plastic material is scratched, it will interact with other abrasive surfaces to wear down the scratched surface itself, as well as any adjacent surface with which it may come into contact.

Plastic products reinforced with abraasive materials can be harmful to humans who handle and work near or with those products. Dermal and respiratory irritation, including possibility of the presence of carcinogenic agents as fillers, are limiting factors for glass fiber and other substances, including wollastanite, and asbestos.

While packaging has been cited as an example, a vast array of other applications could benefit from use of a low cost reinforcing material in a low-end plastic. Typical consumer products that would benefit include boxes, large item housings and other packaging; children's play sets, toys and play houses; luggage shells and reinforcing; bulk packaging including containers and crates used with containerized shipping, home appliances, tools, refrigerators, home furnishings, and furniture; industrial sector packaging of fasteners, parts, pharmaceuticals, food stuffs, manufactured goods, supplies, construction materials, and automotive products; drums for dry goods packaging and storage; boxes; harvesting bins; dry waste bins; household and kitchen products, including storage devices, racks, hangers, liners, and furniture framing and base material; construction/building materials, including wall, ceiling and floor paneling, temporary, interlocking flooring and walkways for buildings and construction sites, permanent, interlocking flooring and roofing for buildings (replacing plywood), shaped, reusable interlocking forms for concrete work, molded joints, connectors, pieces for housing and construction, "work pieces" for building and construction, reusable, movable work benches, saw horses; highway devices including signposts, barriers and molded crash containers; automotive parts including under-the-hood housings and molded interior trim material. Ideally, for injection molded parts, a single component should not exceed the shot capacity of the injection molding machine being utilized to produce the structurally enhanced article. Parts manufactured using continuous extrusion processes, multiple shot/batch press, vacuum or rotational forming, and preformed mat material can, however, be very large structures.

Natural fibers have been considered as strengthening agents for plastics, despite their polar, hygroscopic, hydrophilic characteristics. Of the fibers considered,

jute-like fibers such as jute, kenaf, roselle, aramina and sunnhemp (*Crotalaria juncea*) have the best combination of low cost and high strength. The long cellulosic fibers of jute-like plants are the world's most abundant natural fiber source, after cotton. Fibers from jute, kenaf, roselle, aramina and sunnhemp share similar characteristics. Table 1 presents a typical fiber profile:

**Table 1: Typical Jute-Like Plant Fiber Composition & Characteristics**

<b>Cellulose</b>	<b>55% - 65%</b>
<b>Hemicellulose</b>	<b>20% - 26%</b>
<b>Lignin</b>	<b>12% - 16%</b>
<b>Specific gravity:</b> (48% to 56% of glass fiber)	<b>1.2 - 1.4</b>
<b>Specific modulus:</b> (116% to 166% of glass fiber)	<b>35 to 50 GPa</b>
<b>Tensile modulus:</b>	<b>60 to 120 Gpa</b>
<b>Tensile strength:</b>	<b>10 to 20 Gpa</b>
<b>Diameter:</b>	<b>10 to 20 <math>\mu</math>m range.</b>
<b>length:</b>	<b>2 to 10 mm</b>
<b>Aspect ratio:</b>	<b>100 to 1000</b>

Jute-like fibers grow naturally in long "fiber bundles" or strands that can exceed 3 meters in length. Fiber bundles are used in the manufacture of threads and twine, which have been heretofore the principal applications of these fibers. Unbundled fibers in which lignin and hemicellulose bonds between individual fibers are mechanically and/or chemically broken down are typically used in the manufacture of paper and other pulped products. Individual fiber bundles typically have a tensile strength and tensile modulus approximately half (50%) that of individual fibers.

With regard to ligno-cellulosic composites generally, a significant number of



references exists in this area of art. These references include:

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The focus, with respect to getting natural fibers into plastic, has been that of trying to get the longer fibers into an extrusion. For example, U.S. Patent No. 5,213,889 to Cogswell et al, issued on May 25, 1993 to Imperial Chemical Industries, and entitled "FIBRE-REINFORCED COMPOSITIONS AND METHODS FOR PRODUCING SUCH COMPOSITIONS," discloses the problem

of pulling long fibers through viscous molten resin while achieving adequate wetting. This process is known generally as pultrusion.

The methods disclosed by Cogswell et al were specifically for continuous fibers where the fibers were sufficiently long to give a roving or tow of sufficient strength under the processing conditions used, to be hauled through the molten polymer without the frequency of breakage which would result in an unworkable process or resulting product. Fiber alignment is an important feature of the pultrusion process. This method of Cogswell et al is so narrowly defined, that the reference teaches that fibrous products such as mats made up of randomly disposed continuous fibers are not suitable for use in the invention unless they form part of a fiber structure in which at least 50% by volume of the fibers are aligned in the direction of the draw.

This pultrusion process has its own specialized drawbacks, including the necessity to apply an electrical charge to the bundles of fibers to spread them as they are being wetted by the molten resin. Other methods were contemplated in Cogswell et al to maximize contact between the fibers and the plastic material, including the extrusion of both through a die, and the pressing of both through rollers. All of the above methods make clear that the central idea is such that the longitudinal continuity of the fiber orientation cannot be broken. Further, Cogswell et al required the tensioning of the fiber bundles, which combined with the above considerations makes for a difficult and onerous process.

Cogswell et al go on to describe a utility for the product of his process where the resulting wetted fibers are chopped into pellets or granules where the fibers will have a length of no less than 3.0 millimeters to 10.0 millimeters. Under circumstances where bonding with the surrounding matrix is less than adequate, the very long

aspect ratios that can be achieved with fiber bundles will result in a final composite having superior strength characteristics to that produced with individual fibers. Early attempts at using jute-like fibers to reinforce thermoplastic composites have utilized long bundled fibers and formed the material using thermal presses.

Jute-like fibers are hygroscopic and inherently polar. This renders them incompatible with thermoplastics that are non-polar and hydrophobic. Inter facial sheer properties between fiber and matrix are inherently weak. Results have been disappointing.

Early attempts to compatibalize jute-like fibers for blending with synthetic fibers have experienced some success using pretreatment with a variety of agents, including acrylic acid, sodium hydroxide, acrylonitrile, acrylamide, methyl methacrylate, permanganate, silane, titanate and tolulene diisocyanate. The strategy, in most cases, has been to graft copolymerize with the abundant hydroxyl groups on the surface of jute-like fibers. Again, results in this area have been disappointing. What is therefore needed is a low cost reinforcer and low-end plastic composite which would hold both cost and performance advantages for all packaging presently involving use of plywood or composite board sheeting and wooden boards.

U.S. Patent 5,145,891 issued to Ichiharashi et al on September 08, 1992 and entitled "POLYPROPYLENE RESIN COMPOSITION" describes a composition formed by blending a predetermined amount of organic fibers with a propylene-ethylene block copolymer having a 3-16% by weight ethylene content. The composition is described as modified (0.05% - 15% by weight) by an unsaturated carboxylic acid or its derivatives, acrylic acid, methacrylic acid, maleic acid, itaconic acid, fumaric acid, citraconic acid, esters of those acids, maleic anhydride, citraconic anhydride, itaconic anhydride, acrylic anhydride and methacrylic anhydride. A

preference is given to a glycidylester of methacrylic acid and maleic anhydride. No mention is made of the rationale for inclusion of the modifier, and the range of potential use (0.5% to 15%) is massive. The authors also fail to describe an intermediate step of the present invention that requires manufacture of a grafted polyolefin. This is critical to achieving a bond of maximum strength between lignocellulosic material such as jute, and the surrounding copolymer matrix. The "maleated polypropylene" produced in this manner is optimized for high molecular weight and reactivity. Connection with the surrounding co-polymer matrix is achieved through molecular entanglement (hence the value of the long chain), while the two carboxyl groups of the maleic anhydride react with the OH groups of the jute-like fiber "backbone." This mechanism of attachment is significantly different than the one achieved by the invention of Ichiharashi et al..

Ichiharasi describe the composition as having high rigidity and demonstrating significantly less warpage than moldings of neat polypropylene or propylene-ethylene co-polymers.

While Ichiharsi et al. use only cotton, glass and nylon fibers in the examples cited in the patent, they speculate that jute fibers, among others, including "polyamide fibers, polyester fibers, polyvinyl alcohol type fibers, polyvinylidene type fibers, polyethylene fibers, polyacrylonitrile type fibers, carbon fibers, phenol type fibers, rayon fibers, acetate fibers, flax fibers, ramie fibers, wool fibers, and silk fibers," and any mixture of those fibers can be used. They make no distinction between "natural" or ligno-cellulosic fibers and extruded synthetic fibers, placing them all within a very broad and general category of "organic fibers." Ichiharsi et al. state a preference for polyamide type fibers, polyester type fibers and cotton fibers in some mixture and in any configuration - ". . . woven, knitted or non-woven . . ." Fiber content is preferred

at between 30% and 50%, by weight, of the final composite.

Ichiharasi et al. describe a related composition which also contains an amount of inorganic filler which at its maximum amount should be less than 20%, by weight, of the final composition.

Ichiharasi et al. describe production of the "composition" as involving heating, stirring and kneading in a Heschell mixer or super mixer at 190°C or by melting and kneading in "a banbury mixer, a roll, a single screw or twin screw extruder or co-kneader." This should be followed by production of pellets. The inventors state that care should be taken to prevent fiber decomposition and recommends practices wherein the "original shape as fibers are maintained." They conclude by recommending that the final composition can be molded using any conventional plastics forming apparatus.

Ichiharasi et al. suggest that the composition may prove to be a superior replacement for glass-fiber reinforced material in automotive interior trim, because it has demonstrably less deformation under heat stress than do products manufactured from that material.

Ichiharasi et al. pay little attention to manufacturing processes, stating only that a virtual grab bag of fiber types and one of a variety of unsaturated carboxylic acid acids may be mixed with an ethylene-propylene copolymer in a variety of commercially available equipment (banbury, single screw, twin-screw among others) for an indeterminate amount of time. The method they describe, however, cannot be exercised on conventional equipment, due to the inherent volatility of the carboxylic acids (and their derivatives) they prescribe. To do so in a controlled atmosphere chamber would be prohibitively expensive. As such, the processes - and the almost infinite variety of compositions that may result from those processes - have no

practical application.

### Summary of the Invention

The specific advantage of the composite material formed by the claimed process is that it enables manufacture of a wide range of recyclable structural and packaging materials that have superior strength-for-weight and lower cost than comparable wood-based and polymer-based products. With respect to wood-based products, the thermoplasticity of the composite material enables manufacture of structural forms that have superior strength, utility and safety. For example, from a utility standpoint, a pulp-paper based drum cannot be formed in a tapered shape, and the plywood box cannot be molded to include integral fasteners and strength-optimizing ribbing. Note that with respect to safety, the composite box will not chip - as do plywood or wood boxes and crates. With respect to comparable plastic materials the products of the present invention have lower cost, as well as superior strength and creep attributes. With respect to comparable conventionally reinforced plastic products the composite-manufactured materials have similar strength and creep attributes, but enjoy significant strength for weight and cost advantages. These advantages can be competitively applied to a wide range of industrial and consumer applications.

On a more fundamental level, the solution involves the grafting of a pendant chain of plastic to the fiber which is compatible with the encapsulating thermoplastic matrix. Maleic anhydride is used to react with the lignin and the cellulose of the fiber, producing a stable, high strength bond. Another general objective is to maximize the fiber aspect ratio. This translates into seeking to minimize agitation during blending to achieve a maximum strength bond. The long fiber length (up to 500 aspect ratio),



results in high strength. Additional stiffness can be imparted with the addition of microsphere and powdered organic filler material (such as powdered kenaf and jute core material) without sacrificing strength.

In addition to maleic anhydride, acrylic acid and its derivative anhydride have also been successfully used in the textile industry. The OH groups on the "jute, kenaf, roselle, crotalaria or aramina backbone" - react with carboxyl groups in acrylic (one) or maleic anhydride (two) - called graft copolymerization. Another beneficial substance is the use of recently available maleic-anhydride-grafted polyolefins as compatibilizers. This has resulted in considerable enhancements in the properties of the composite of the present invention. Finally, inclusion of a small amount (<5%) of unreacted thermoset polymer, particularly, phenolic or epoxy resins (and their catalysts) in the final molding process produces additional property enhancements through dynamic cross-linking, than are achieved by maleic anhydride grafting alone. The present invention also enhances the convenience to the molding industry, thereby also reducing their costs, by incorporating all compound ingredients, including the thermoset resins, in a single cold-pressed pellet.

The present invention utilizes maleated polypropylene to form strong anhydride-hydroxyl bonds through the reaction of the two carboxylic groups of maleic anhydride with the hydroxyl groups on the fiber surface. The resulting interfacial shear is superior to that of other known graft copolymers. Grafting a pendant chain to the fiber that is compatible with the encapsulating thermoplastic matrix. Maleic anhydride reacts with the lignin and the cellulose, producing a stable, high strength bond. The strength of the maleated polypropylene-fiber bond is principally a function of the percentage of maleic anhydride in the maleated polypropylene.

Connection with the surrounding polymer matrix is through molecular

entanglement. The strength of the connection is a function of the length of the maleated polypropylene molecular chain and the crystallinity of the surrounding matrix.

The present invention also allows discretionary use of dynamic cross-linking with phenolic and epoxy resins to achieve enhanced composite material strength. Resin and catalyst application must coincide with the bonding of maleated polypropylene with fibers.

Preferably, preparation of the composite material can be accomplished by the steps of preparation of the bast fiber by either retting or mechanical separation of fiber bundles, and preparation of the transported bast fiber for processing - involving combing to "open up" baled material and discretionary chopping to reduce fiber bundle lengths to between 1 and 20 centimeters. Next, the maleated polypropylene is prepared. If commercially marketed maleated polypropylene appropriate to the specific application is not available, then maleated polypropylene should be manufactured by mixing maleic anhydride (3% to 6%) with powdered polypropylene or finely ground polypropylene pellets or particles. The size of the particles or pellets may vary, but the finer or smaller the size of the granules or pellets, the better the result. Preferably the mean diameter of the pellets will be below 10 microns. The reaction is catalyzed with 3% by weight t-butyl peroxyester and conducted for 2 hours in an inert atmosphere at a temperature of from about 120° to about 130° C.

Next, the fibers and up to 20% by weight maleated polypropylene (and possibly phenolic resin and hexamethylene tetramine at up to 5% and 0.5% by weight of the final composite respectively, or partially reacted phenolic resin inclusive of hexamethylene tetramine at up to 5.5%) are blended in such a manner as to achieve even coating of the fibers with an intended amount of maleated polypropylene (and

phenolic resin). This blending can be achieved using several techniques, described more fully in the detailed description. Lastly, the maleated polypropylene/resin-coated fiber (20% - 60%) is blended with polypropylene (80% to 40%) and pelletized for subsequent molding or directly applied a molding molding process. While the material is particularly suitable for injection molding, it may also be press molded, extruded, rotation molded, blow molded or vacuum formed on various conventional molding apparatus. The resulting molded composite natural jute-like reinforcing fibers are matrix sensitive - as opposed to surface sensitive - and therefore, unlike glass, will not shear as easily during molding during injection or extrusion. Theoretically, Strain from load is transferred through the resin mix from fiber to fiber. The fiber is stronger and has a higher modulus than the matrix. Much higher stress is induced in the fiber than is carried in the matrix surrounding it.

#### Brief Description of the Drawings

The invention, its configuration, construction, and operation will be best further described in the following detailed description, taken in conjunction with the accompanying drawings in which:

Figure 1 is a process flow diagram which illustrates the overall process steps possible regarding the composite and method of making of the present invention;

Figure 2A illustrates the shape of the wall sections into which the composite of the present invention was made, in order to perform further testing;

Figure 2B illustrates the test setup with which the wall sections of Figure 2A were tested;

Figure 3 is a graphical comparison of tensile stress-strain curves for the blends of Example 1;

Figure 4 is a bar graph which compares notched impact energy for each of the

composite blends of Example 1;

Figure 5 is a bar graph which compares the un-notched impact energy for each the composite blends of Example 1;

Figure 6 is a bar graph which compares the cantilever modulus of elasticity for each of the composite blends of Example 1;

Figure 7 is a bar graph which compares the maximum cantilever strength for each of the composite blends of Example 1;

Figure 8 is a bar graph which compares the flexural modulus of elasticity for each of the composite blends of Example 1;

Figure 9 is a bar graph which compares the maximum flexural strength for the composite blends of Example 1;

Figure 10 is a bar graph which compares the tensile modulus of elasticity for the composite blends of Example 1;

Figure 11 is a bar graph which compares the maximum tensile strength for the composite blends of Example 1;

Figure 12 is a bar graph indicating the change in mass after water soak of each of the composite blends of Example 1;

Figure 13 is a bar graph indicating the percent change in length after ambient water soak of the composite blends of Example 1;

Figure 14 is a bar graph indicating the percent change in width after ambient water soak of the composite blends of Example 1;

Figure 15 is a bar graph indicating the percent change in thickness after ambient water soak of the composite blends of example 1;

Figure 16 is a bar graph indicating the percent change in cantilever secant modulus after ambient water soak of the composite blends of Example 1;

Figure 17 is a bar graph indicating the percent change in maximum cantilever strength after ambient water soak of the composite blends of Example 1; and

Figure 18 is a graph of the generalized relationship between jute and kenaf fiber loading and composite strength for fiber lengths of one millimeter and one centimeter.

### Detailed Description of the Preferred Embodiment

There are a number of advantages to using natural fibers as reinforcing material in polymeric composites. Major factors include low cost, low weight, and abundance. But in addition, natural fibers have the advantages of high specific strength and stiffness (property value divided by weight), and low abrasion to processing equipment, compared to typical mineral and glass reinforcing materials. processing technology. The matrix sensitive fibers are, unlike glass fibers, amenable to multiple recycling episodes without resulting in significant loss of strength in molded articles manufactured from the recycled material. Finally, the present invention allows use of low-cost, low-end thermoplastics such as polypropylene and high density polyethylene (HDPE), in combination with discrete binders and compatibilizers, to compete favorably against high-end polymers and more expensive synthetic fiber (glass, nylon, kevlar, boron, carbon etc..) reinforced polymers.

While a number of recent articles have reported favorable material properties of natural fiber/thermoplastic composites, and projected the feasibility of such systems to be produced on a commercial basis. Until now, only a small amount of research has been done on jute, kenaf and other jute-like bast fibers as reinforcements in extruded and/or molded thermoplastics. Jute, kenaf and roselle are known to be some of the strongest natural fibers, and have been used for centuries in a variety of

applications, including twine, rope, carpets, fabric and pulp-based applications.

These "jute-like" fibers also offer an inexpensive, yet strong, alternative to mineral fillers or glass fibers as reinforcers for low-end thermoplastics. The term "jute-like" includes jute, kenaf, roselle, aramina, and sunnhemp (*crotalaria*).

Thermopolymer material that can be utilized as the base matrix material in the present invention may have a material density ranging between about 0.908 and 1.08 g/cm<sup>3</sup> and have a melt flow index (MFI) of between 8 and 50 g/10 min at (230°/2160 g). Elongation at break of the resulting composite material is less than about 8% with respect to the decrease in impact strength. At a density of approximately .9 g/ml, the melt flow index will range between 8 and about 50 g/10 min (230°/2160 g). The composite material of the present invention is more ductile than brittle.

In a preferred formulation, the present invention utilizes maleic anhydride in combination with polypropylene and natural jute-like fibers to form a reinforced, maleated thermoplastic. Specifically, maleated polypropylene is employed to form strong anhydride-hydroxyl bonds through the reaction of the two carboxylic groups of maleic anhydride with the hydroxyl groups on the fiber surface. The resulting interfacial shear is superior to that of other known graft copolymers. The strength of the maleated polypropylene-fiber bond is principally a function of the percentage of maleic anhydride in the maleated polypropylene.

Connection with the surrounding polymer matrix may be predominantly through molecular entanglement. The strength of the connection is a function of the length of the maleated polypropylene molecular chain and the crystallinity of the surrounding matrix and the fiber aspect ratio.

The present invention also allows discretionary use of dynamic cross-linking

with phenolic resins to achieve enhanced composite material strength. Resin and catalyst application should coincide with the blending of maleated polypropylene with fibers. Although the contemplated variations are many, preparation of the composite material is centrally related to the following discrete steps.

First, the fiber is obtained. As a background, jute-like fibers are obtained from harvesting on an annual crop basis. The plant material of interest is the external bast which are bark based fibers of the jute-like plant. The fiber characteristics include strong polarity and a relatively lower percentage of lignin and hemicellulose than other natural lignocellulosic fibers, therefore making it a potentially a better binder.

**Table 2: Approximate Composition of Jute-Like Fibers**

<b><math>\alpha</math>-Cellulose 61% crystalline</b> (This is a linear polymer of $\beta$ -D-glucose)	
<b>Hemicellulose</b> <b>Amorphous</b> (These are low molecular weight polysaccharides)	<b>23%</b>
<b>Pentosans</b>	<b>16%</b>
<b>Ash</b>	<b>1%</b>
<b>wax</b>	<b>1%</b>
<b>Nitrogenous matter</b>	<b>2%</b>
<b>Acetyl value</b>	<b>3.5%</b>
<b>Polyuronide</b>	<b>5%</b>
<b>Lignin</b> (amorphous - polyphenolic)	<b>14%</b>
<b>Specific gravity</b> (2.5 for Glass)	<b>1.29</b>
<b>Specific modulus</b> (30 Gpa for Glass)	<b>40 Gpa</b>
<b>Tensile Strength</b>	<b>12 Gpa</b>
<b>Tensile modulus (kenaf)</b>	<b>60 Gpa</b>
<b>Flexural Strength (fiber bundle)</b>	<b>116.3 MPa</b>
<b>Flexural Modulus (fiber bundle)</b>	<b>6.63 GPa</b>
<b>Jute-like fiber diameters</b> <b>range.</b>	<b>10 - 20 <math>\mu</math>m</b>
<b>Jute-like fiber lengths</b>	<b>2 - 3 mm</b>
<b>Aspect ratios ranging</b>	<b>100 to 300</b>

The jute-like fibers are lignocellulosic fibers which are the most abundant renewable agricultural fiber resource, after cotton. The fibers are typically obtained in filaments comprising bundles of individual fibers. Bundles can be up to 3.5 meters long, but have a tensile strength and modulus approximately half that of the individual fibers. Individual fibers, themselves, can have aspect ratio's between 100 and 500 (ratio of length to width) and are very efficient reinforcing agents. A 10 fold increase in aspect ratio will result in an approximate doubling of tensile strength - which, nevertheless, derives its primary power from the tensile strength of the underlying fiber. Other differences with respect to glass fibers, aside from cost, include energy processing requirements. Jute-like fiber processing consumes only 2% of the energy used to produce and process glass fiber.

The use of these annual-growth lignocellulosic natural fibers as reinforcing fillers is appealing, both because of the properties of the resultant polyolefin composites and from an environmental viewpoint. Agro-based natural fibers, with their high aspect ratios and high specific properties, have the potential to be outstanding reinforcing fillers in thermoplastics.

The nonabrasive nature of jute-like fibers permits the use of high fiber loadings without the extensive damage to compounding and molding equipment that can occur with much harder mineral-based fibers/platelets such as glass and mica.

The high moisture absorption of all natural fibers and their low microbial resistance are disadvantages that need to be considered, particularly during shipment and long-term storage. These disadvantages are minimized in composites such as those resulting from the present invention because individual fibers are completely encapsulated by the surrounding polymer matrix. Nevertheless, care should be taken to select applications where these potential drawbacks do not hold great significance.



If necessary, moisture absorption can be significantly reduced through the acetylation of some of the hydroxyl groups present in the lignocellulosic fiber, although at some increase in cost. Some of the major pros and cons of lignocellulosic fibers as reinforcing fillers are listed below in Table 3.

**Table 3: Advantages and Disadvantages of Lignocellulosic Fibers**

<u>Advantages</u>	<u>Disadvantages</u>
<ul style="list-style-type: none"> <li>• Low cost</li> <li>• Renewable</li> <li>• Low densities</li> <li>• Nonabrasive</li> <li>• Biodegradable</li> <li>• Low energy consumption</li> <li>• Reusable-recyclable</li> <li>• Good strength retention on reuse</li> <li>• High specific properties</li> </ul>	<ul style="list-style-type: none"> <li>• High moisture absorption</li> <li>• Poor microbial resistance</li> </ul>

Jute, kenaf, sunnhemp (crotalaria), roselle, and aramina fibers, among the category of jute-like fibers, may all be utilized with the present invention. Kenaf fibers are extracted from the bast of the plant *Hibiscus cannabinus*, and filament lengths of about 1 m are common. The filaments consist of discrete individual fibers, generally 2 to 6 mm long, which are themselves composites of predominantly crystalline cellulose, amorphous cellulose, lignin, and hemicelluloses. Fiber properties can vary depending on the source, age, separating techniques, and history of the fibers. It is preferred that individual fibers remain chemically bound as filaments (fiber bundles) because pulping procedures needlessly consume significant energy resources. Among jute-like fibers, kenaf holds particular importance because it is

now a crop grown commercially in the United States. The use of more expensive, higher strength, or longer aspect ratio agro-based fibers, such as cotton, flax, ramie, and hemp may result in some better properties than those obtainable with jute-like fibers. None, however, can compete with with jute-like fibers with respect to the cost-efficiency of those properties. Processed jute and kenaf fibers, for instance, can be obtained for less than one third the price of flax and one seventh the price of cotton, yet they produce comparable strength properties when employed as reinforcers in composite materials.

Jute is a strong coarse fiber used for making burlap, gunny sacks, cordage and carpet material. It is obtained from two east India plants, *Corchorus capsularis* is and *Corchorus olitorius* of the linden family. These fibers are important with respect to the present invention because of their relative abundance and low cost.

Once obtained, the jute-like bast fiber is prepared by either retting or mechanical separation of fiber bundles. Cleaned, separated fibers are then either baled for transporting, or directed to a shredder/chopper, where fiber bundles or filaments are reduced to lengths to between 1 and 20 centimeters.

Next, maleated polypropylene is prepared. If a commercially marketed maleated polypropylene appropriate to the specific application is not available, then maleated polypropylene should be manufactured by mixing maleic anhydride (3% to 6%) with powdered polypropylene. The reaction is catalyzed with 3% by weight t-butyl peroxyester and conducted using slow mixing for 2 hours in an inert atmosphere at 120° - 130° C. The result is the maleated polypropylene compound used as a binder in the present invention. Specific properties of the maleated polypropylene are a function of maleic anhydride strength and amount, catalyst amount, reaction time and temperature and polypropylene characteristics - principally

molecular weight and length of the pendant chain.

Next, the jute-like fiber material and up to 20% maleated polypropylene is blended in such a manner as to achieve even coating of the fibers with an intended amount of maleated polypropylene. As an option, it is possible to blend in phenolic resin or epoxy resin in amounts which can be calculated to yield up to approximately 5% by weight of the final composite. A preferred amount of phenolic resin or epoxy resin in the final compound will range between 0.5 and 1.5% by total weight. Phenolic resin is preferably applied in a semi-reacted state as a fine powder, but appropriate amounts (by supplier recommendation) of hexamethylenetetramine (hexa) curing agent can also be blended directly during the final molding process. The blending step can be accomplished by mixing, charged particle, free falling contact application, or dipping.

Where blending is a vigorous mixing step, the mixing of fiber and blended, powdered maleated polypropylene and semi-cured phenolic resin and/or phenolic resins and hexamethylenetetramines and/or epoxy resins and catalysts is accomplished in an air-lay or pneumatic process. A continuous bat or mat of coated fiber can then be passed through an infrared oven or hot rollers to fix the powders. This step is preferably performed at a temperature ranging between 160° C to 180° C, and preferably at about 170° C.

Where blending is a free-fall step, the, powdered maleated polypropylene, and semi-reacted phenolic resins and/or phenolic resins and catalysts and or epoxy resins and catalysts may be electrostatically charged and spray-attached to the chopped, separated fibers in a free-fall reaction chamber. The coated fibers are then passed by belt through an infrared oven as described in the mixing step to the same temperature range.

Where blending is a dipping step, the maleated polypropylene is applied to the fiber by dipping a continuous bat or mat of fiber through an emulsion bath. The emulsion preferably consists of maleated polypropylene at between 20% and 40% by weight, water content of from about 50% to about 70% by weight, potassium hydroxide of from about 2% to about 4% by weight, sodium metabisulfite in an amount of about 0.3% by weight and an appropriate surfactant in an amount of from about 5% to about 8% by weight. The surfactant is preferably one such as Igepal CO-630, which is commercially obtainable from the Rhone-Poulenc, Prospect Plains Road, Cranbury, NJ. 06512. Igepal CO 630 is a nine mole ethoxylate of nonylphenol, which will be referred to simply as nonylphenol. It is made by reacting ethylene oxide (oxirane) with nonylphenol. The dipping method is not suitable for inclusion of phenolic or epoxy resin components.

While the electrostatic charged powder application is generally preferred, the airway or pneumatic process is acceptable, and the dip method is suitable for lower molecular weight maleated polypropylene. Again, the maleated polypropylene modifies the degree of adhesion - or interfacial shear properties between the matrix and the fiber. The bat or mat is, as was the case with mixing and free-fall, air dried in an infrared or other type oven at the temperatures recited above for the mixing step, before being passed through hot rollers for fixing.

The combined fiber and maleated polypropylene (and possibly phenolic or epoxy resin) mixture, having been heat-fixed is then further blended with the primary polymer, preferably filamented polypropylene of between 8 and 50 g/10 min. MFI, and comprising between 40% and 80%, by weight, of the final composition. This can be achieved by various means, the preferred method being by a mixing airstream wherein ingredients are free-mixed during carriage to a cold-press pelleting

apparatus. There, the blended ingredients are directed by the airstream to the face of the die and forced by the pelletizer's pressing/idler wheels through a die, wherein pressure and heat generated at the die surface cause a skin to form on the material perimeter. This further causes the ingredients to bind together in such a manner as to form discrete pellets. Polypropylene in the form of flake, powder, pelleted or ground material is also acceptable.

There are still other, acceptable, but less effective blending techniques that involves melting of the carrier polymer. In one method, the ingredients are introduced simultaneously and batch mixed in a vessel such as a Banbury Mixer or a Farrel Continuous Mixer (FCM). Mixing occurs when the base polypropylene melts due to shear induced temperature elevation. Once the mixture is completely fluxed, mixing should continue for an additional 30 seconds to 1 minute to enable adequate blending. The fluxed mixture can then be fed through standard extrusion pelleting apparatus to form pellets.

In another acceptable, though less preferred mixing method, measured, balanced, continuous streams of ingredients, including the maleated polypropylene/fiber mixture and polypropylene, as either filaments, powder, pellets, flakes or ground material, are fed directly through a twin or single mixing screw, wherein the material is shear-melted and mixed, and from whence the material is directed to pelleting apparatus to form pellets.

It should be noted that in any instance where an intermediate step of manufacturing fluxed, or melt-formed pellets is contemplated, the mixture may not include either phenolic or epoxy resins and their associated catalysts. The reasoning behind this is that the thermoset polymers would react during fluxing and pellet formation, thus nullifying their utility during subsequent injection or other molding

processes.

Mixing that involves use of an apparatus that generates temperature elevations through shearing action, will generally result in damage to fiber that exceeds that of other preferred methods.

A third and less preferred category of mixing methods precludes the stage of first blending maleated polypropylene (and possibly phenolic and epoxy resins) with the jute-like fibers. This method includes simply adding all ingredients simultaneously in either: (a) a mixing airstream and cold press pelleting operation; a batch banbury or FCM process followed by extrusion-pelletizer; or (c) a twin or single screw mixing extruder and pelletizer. The results (compound quality) produced by these methods differ from preferred methods (see above) in that the statistical probability that maleated polypropylene will (be able to) react with jute-like fiber surfaces are significantly lower. This is presumed to result in marginally inferior mechanical properties for parts, and equipment manufactured from these compounds.

The issue in the use of maleic anhydride is the "interphase properties" of the substance. One key aspect is high molecular weight for the polypropylene and high percentage of maleic anhydride. The method of connection with the fiber is due to maleic anhydride (covalent acid-based interaction - formation of anhydride-hydroxyl covalent bonds) and the connection with the polymer is through molecular entanglement with the surrounding polymer matrix. Note that the more amorphous the polypropylene - i.e., the less crystalline - the longer the molecular chain should be - or, the greater the molecular weight.

Maleated polypropylene contains succinic anhydride moieties which are located at the ends of the polypropylene chains. The polar maleic anhydride is

strongly attracted to the polar fiber surfaces. In one-step mixing, despite being less than .02% of the composition, a significant percentage of the maleic anhydride interacts with the fiber surface. Two carboxylic groups of maleic anhydride react with the hydroxyl groups on the fiber surface.

Once the fibers have been initially introduced to the maleated polypropylene, as described above, to form the maleated polypropylene/resin-coated fiber preform, it is added to polypropylene for final mixing to form the composite of the present invention. The fiber preform is added to the polypropylene in a ratio of from about 20% to about 60% preform to about 80% to about 40% polypropylene. Once this final composition is formed, pelletizing will aid in handling and later use for injection molding, press molding, rotation molding, blow molding, vacuum molding and/or extrusion. Of course, the final material composition may be directly injection molded, press molded, rotation molded, extruded, press-formed, vacuum molded or otherwise used immediately upon its formation.

The final mixing of the maleated polypropylene/resin-coated fiber preform with the polypropylene (or polyethylene) to form the final composite composition may, as has been described above, be accomplished by several methods. The preferred method of mixing and pelletizing can be accomplished with the use of a specially adapted cold-press pelleting mill, sometimes also known as the California Pellet Mill - after the American company of the same name (A number of other manufactures do, however, make similarly configured pelleting machines). This system uses a drum die rotated in a vertical plane (using a horizontal drive). Stationary, opposing compacting compression wheels ride inside the die. The material is force fed, usually within an airstream, through a front opening against the inner face of the die. With this preferred configuration, there is less grinding of the

material than with vertical axis, horizontal plane rotation, flat dies.

Another variation on this method is sometimes known as the Kahl process - after the German company of the same name (a number of other manufacturers do, however, make similarly configured pelleting machines). This device pushes material through a die, which is usually horizontally disposed, by using grooved metal wheels travelling about a vertical axis. The material to be treated is usually gravity fed from above the Kahl system. The outer edge of the Kahl die maintains a higher rotational velocity than the inner, active portion of the die, and thus introduces significant grinding to the material being treated. While grinding is useful in the treatment of some substrates, it is detrimental to the jute-like fibers processed in this fashion.

Both of these systems form pellets by pushing material through holes drilled through the dies. Pellet integrity is maintained as a function of compaction and strength of the outer "skins" formed by the process. The strength of the skin is a function of the materials employed, die configuration and the maximum temperature achieved during the process. Conversely, the maximum temperature is a function of the material density, toughness and abrasiveness, hole diameter and taper, hole distribution and die thickness. With the California Pellet Mill, a smoother and more uniform product is formed, as compared with the Kahl machines. Pellets manufactured using the California Pellet Mill process also produce a molded product having superior strength characteristics to those manufactured on Kahl-type machines. Therefore, as between these two types of machines, the California Pellet Mill is the preferred mode. With either machine, the pellet size can be controlled by mounting a blade in a position to "shave" the material as it exits the die holes.

In a less preferred method, measured quantities of maleated polypropylene/resin- coated fiber preform are added to a melted batch of



polypropylene and mixed at medium speed for between 5 and 10 minutes. The favored method of batch mixing is with a mixer such as a Banbury and Farrel Continuous Mixer. These mixers use the shear process to create heat, melting the polypropylene (or polyethylene) and thus assisting in physical mixing. Once mixed, the final material composition is passed to an extruder and pelletizer or directly to an injection (or other) molding machine.

It is noteworthy that the cold press pelleting method for mixing and pelleting the compound ingredients enjoys significant advantages over other, less preferred methods. Specifically, these advantages include: (a) the ability to include phenolic and epoxy resin constituents in an un-fluxed (unmelted) pellet; and (b) significant cost advantages amounting to a final pellet production cost of between one eighth and one third that of other methods.

The final acceptable, though less preferred mixing method, involves feeding a measured, balanced, continuous streams of ingredients, including the maleated polypropylene/fiber mixture and polypropylene, as either filaments, powder, pellets, flakes or ground material, directly through a twin or single mixing screw, wherein the material is shear-melted and mixed, and from whence the material is directed to pelleting apparatus to form pellets.

The next step is then to mold products using prepared pellets or hot-blended compounds under conditions in which material temperatures are maintained below about 230°C and injection pressure, should that method be used, is held below about 28,000 psi. Rheological properties can be improved by selecting a polypropylene having a high melt flow index. A resulting reduction in flexural modulus can be partially compensated by choice of more highly crystalline polypropylene. Fiber orientation, which provides significantly higher directional strength, is achieved by

varying flow convergence and expansion. Injection flow under convergent conditions leads to axial unidirectional orientation, and under expanding conditions leads to orthogonal orientation. The resulting composite material of the present invention is characterized by high stiffness, low creep, high tensile and flexural strength; low weight (relative to other reinforced plastic material) and low abrasiveness (relative to other reinforced plastic material). Products manufactured from the composite material and having between 30% and 60% fiber content (by weight) have tensile and flexural strength of between 2 and 6 times that of similar material having approximately the same overall weight and volume that is manufactured from the underlying neat thermopolymer material (polypropylene and polyethylene).

Preliminary examination of the utility of kenaf and jute bast fibers as reinforcing fillers in melt-processed polypropylene composites, both with and without phenolic and epoxy additives, yields a vast array of potential uses. One proposed use for such composites is a 55 gallon drum for the purpose of storing and shipping foods, chemicals, small manufactured goods and various commodities. Example 1, below, compares composites employing jute and kenaf fibers with comparable sections cut from a 55 gallon rolled paper drum. Jute and kenaf fibers were reduced to a size enabling introduction, along with other composition ingredients (maleated polypropylene and base polypropylene), into a small laboratory-scale high-intensity thermokinetic mixer ("K-Mixer"). The resultant blends were both injection molded into test specimens and formed into standard ASTM test bars or curved cylindrical-sectioned panels for subsequent mechanical property measurements.

Blends of 50/50 by weight kenaf/polypropylene (12 melt flow index) and jute/polypropylene (12 melt flow index) were prepared, with and without coupling agents (maleated polypropylene). In addition, blends of 40/60 by weight

kenaf/polypropylene with the maleated polypropylene mixture coupling agent were prepared using two different types of polypropylene. The polypropylenes were Polypropylene-L (12 melt flow index) and Polypropylene-H (50 melt flow index-nucleated, high stiffness). Typical mechanical tests including stiffness, strength, and Izod impact, along with water absorption, were performed, in addition to a modified compression test. The resulting composite material was compared to existing samples cut from standard wound fiber drum material.

The results indicated that, overall, the kenaf and jute composite material significantly outperformed the drum material in stiffness, strength, and compression, but had lower impact strength values. Jute fiber tended to impart more favorable values in properties than did kenaf fiber, in both mechanical property values and decreased water absorption effects. The effect of high melt flow index polypropylene compared to low melt flow index polypropylene on most mechanical properties was minimal.

An alternative, generalized method of producing the composite of the present invention includes the formation of a molded article. A molded article is formed from a natural jute-like fibre reinforced thermoplastic composition in a process that includes several steps. The steps include: (a) the step of evenly blending a composition containing between 80% and 99% by weight natural jute-like fibers and between 1% and 20% maleated polypropylene as powder, emulsion or melted polymer, (b) the step of heat-fixing a bond between the jute-like fibers and maleated polypropylene; (c) the step of chopping the coated fibers to lengths between 1 and 40 mm; (d) the step of melting and homogenizing a composition containing between 10% and 80% maleated polypropylene-coated fiber and 90% and 20% crystalline or semi-crystalline polypropylene; and (e) a step of injection molding, press molding,

blow molding, rotation molding, or vacuum molding under conditions in which material temperatures are maintained below 230°C and injection or press or vacuum pressure is held below 28,000 psi.

It may be helpful to illustrate an overall process schematic to facilitate the viewing understanding of the processes involved in the present invention. Referring to Figure 1, the Jute or Kenaf 51, polymer 53 and coupling agent 55 are mixed together by dry mixing 57. After dry mixing 57, a melt blending step 59 occurs, and then a granulating step 61. As an option, a colorant 63 step may be included. After granulating 61, either of two steps may occur, including an injection molding 65 step, or a panel pressing 67 step.

For the melt blending 59 step, a 1-L high intensity thermokinetic mixer (K-mixer) was employed for melt-blending the fiber and polymer. This apparatus consists of a cylindrical chamber in which several blades rotate at high speed and impart kinetic energy to solids that are present. When the plastic reaches its melting or softening point, fluxing and mixing begin and the temperature begins to rise very rapidly. Within seconds thereafter, the molten mixture is discarded through a bottom door which is temperature activated. For most systems, total blending time may be from 20 seconds up to perhaps 2 minutes.

The effectiveness of blending the K-mixer can be a function of several parameters, including plastic and filler properties, batch size, starting temperature, blade speed, time, and discharge temperature. The K-mixer is excellent for producing well dispersed systems, but may cause fiber attrition. If the composite material of the present invention is performed at or near the quality level required of drum material, more gentle mixing would be employed to maintain fiber lengths to further improve the material properties.

After melt blending 59, each batch of a particular blend may be granulated and mixed with the other batches of that blend. With regard to injection molding procedures 65, the resultant blends may be injection molded into standard ASTM test specimens using a Cincinnati-Milacron 33-ton reciprocating screw injection molder. It may be preferable to injection mold with a flat temperature profile of about 188°C (370°F), and to utilize injection pressures between about 1000 to about 1300 psi maximum. The mold is typically neither heated nor cooled. Once molded, specimens and remaining blend materials are preferably stored over desiccant in sealed bags at 90°F and 20% relative humidity for 3 days before testing.

A portion of the blends were may also be used to produce curved cylindrical panels for modified compression test as described in the introduction. A graphical view of the test set up used for the panels is shown in Figures 2A and 2B. Panels 71 may be shaped to an eleven inch radius as is shown in Figure 2A. Figure 2B illustrates the use of a pressure ball 73 bearing down upon an upper pressure plate 75 which in turn bears down upon the panel 71 specimen under test 71, which bears down upon a lower pressure plate 77, which is supported by a base 79.

To produce the curved panels 71 for the compression test, one method preferably entails the use of 360 grams of the granulated material to be placed in a mold in a hot press and pressed at 193°C (380°F) at a pressure of about 500 psi for 5 min., then at 1000 psi for 1 min. The final dimensions of the pressed panel are preferably 12" x 12" x 0.138". These panels 71 can be mechanically sawed down to 10" x 10" square pieces for further forming. The panels 71 may then be sawed down to 10" x 10" square pieces for further forming. The panels 71 can then be formed to give a uniaxial curvature equal to that of the a fiber drum, such as a fiber drum having a 27.5 cm radius of curvature. This can be accomplished by placing a single panel on

a stainless steel form of equal curvature, placing a mated form on the top of the panel, and loading approximately 8 kg of mass on the top form to assist its correct forming. The entire apparatus can then be placed in a forced air oven at 150°C for 10 minutes, after which time the curved panel can be removed, then cooled and stored under the same conditions as injection molded test specimens.

### EXAMPLE 1

The objective of the present example is to assess the potential of kenaf and jute bast fibers as reinforcing fibers in a polypropylene matrix. The end-use of such a composite would be a stackable, 55 gallon drum, used for the food shipping industries. The ongoing goal is to create an inexpensive, yet strong and durable material, that can subsequently be recycled into the same or similar types of products.

After an initial literature search, laboratory screening experiments were conducted to establish acceptable processing parameters and to give the researchers familiarity with the materials. Once acceptable conditions were established, a matrix of six blends were produced to examine the effects of the variables: fiber type, percent composition, polymer type, and coupling agent. Two additional blends were produced as a qualitative examination of the effects of colorants, for a total of eight blends. The performance of the composite materials were also compared to the performance of a standard fiber wound drum. The effects of these variables were determined by subjecting the composite and drum material to standard ASTM (American Society for Testing and Materials) tests for static and blending modulus, strength, impact properties, and moisture absorption. In addition, since the finished composite drum should withstand axial forces up to 6,000 lbs when stacked in a warehouse, a modified compression test was developed which is described in the

following paragraphs.

Weingarten and others, have developed test methods and theoretical equations to determine the buckling coefficient, and consequently, the maximum axial load, of thin walled cylindrical and conical shells under axial compression. The axial load on cylindrical and conical shells of varying radii, thicknesses, and lengths of certain materials was determined. From this data, they developed a minimum design criteria equation which could be used with reasonable assurance that the structure will meet performance requirements. Relevant to the tapered containers under consideration, the equation for the axial load at buckling is given by equation 1:

$$\frac{P}{2\pi E t^2 \cos^2 \alpha} = 0.606 - 0.546 \left( 1 - e^{-\frac{1}{16} \left( \frac{l}{t} \right)^2} \right) + 0.9 \left( \frac{r}{l} \right)^2 \left( \frac{t}{r} \right) \quad \text{Equation (1)}$$

Where

- P = total axial load at buckling
- E = Young's modulus of shell wall material
- $\alpha$  = semi vertex angle of cone
- t = shell wall thickness
- l = slant length of cone
- r = radius of curvature of small end of cone.

Therefore, by determining the Young's modulus of the material, the maximum needed axial load, and the dimensions of the final product, a minimum wall thickness can be determined. It should be realized that this is in an idealized situation and other factors may act to decrease the load-bearing capabilities. The wall thickness will typically be over-engineered to compensate for any unforeseen variables.

The jute used for this Example 1 was a 50/50 blend of Indian and Bangladeshi

virgin Tossa Grade A Cuttings. Cuttings were taken from the lower stalk section of the jute plant, and ranged from 7 to 12 cm in length. The jute was obtained from Dixie Manufacturing Company of Norfolk, Virginia, as separate bales of the Indian and Bangladeshi fiber. Dixie Manufacturing is a producer of non-woven jute products. The kenaf was obtained from Kenaf International of McAllen, Texas, and was approximately 90 percent by weight bast fiber and 10 percent core, as received. The kenaf fibers were about the same length as the jute fibers and both were washed before processing to remove dirt and core material. The fibers were then dried at 105°C, and then sent through a small granulator to chop the fibers to approximately 1/2" lengths to allow further processing during the melt-blending stage.

Two types of polypropylene were used in this Example 1. The first was a virgin homopolymer in pelleted form obtained from Solvay Polymers (Houston, Texas) - Fortilene 1602. Fortilene 1602 (Polypropylene-L) is a typical injection molding grade polypropylene with a Melt Flow Index (MFI) of 12 g/10 min. FORTILENE 1602 is a polypropylene homopolymer produced by SOLVAY Polymers utilizing unique catalyst technology. It is designed for injection molding and general purpose extrusion. This material is advertised as meeting the Food and Drug Administration requirements of 21CFR 177.1520.

The second was Fortilene 1986 (Polypropylene-H), a controlled rheology homopolymer polypropylene, which is nucleated to promote higher crystallinity. Fortilene 1986 has a higher melt flow index of 50 g/10 min. It is specifically designed for high speed injection molding. FORTILENE 1986 is produced by SOLVAY Polymers utilizing unique catalyst technology. It is both antistated and nucleated and is designed for applications that require improved impact. This material is also advertised as meeting the Food and Drug Administration requirements of 21CFR



177.1520. Property data sheets on these polypropylene's as provided by the

**Table 4: FORTILENE 1602 Polypropylene Data**

Property	ASTM Test	Typical Values	
		English Units	SI
Units	Method		
Density	D 1505	—	
.900 g/cc			
Melt Flow Index <sup>1</sup>	D 1238	—	
12.0 g/10 min			
Tensile Strength			
@ Yield (2 in/min)	D 638	4800 psi	
33.1 MPa			
@ Break (2 in/min)	D 638	4800 psi	
33.1 MPa			
Elongation			
@ Yield (2 in/min)	D 638	8%	8%
@ Break (2 in/min)	D 638	570%	
570%			
Tensile Modulus <sup>2</sup>	D 638	140,000 psi	
965 MPa			
Flexural Modulus <sup>2</sup>	D 790	180,000 psi	
1241 MPa			
Notched Izod Impact Strength			
@ 23°C	D 256	0.3 ft-lbf/in	
Unnotched Impact Strength			
@ 23°C	D 256	12 ft-lbf/in	
640 J/m			
Hardness (Shore D)	D 2240	68	68

(6/19/91)

<sup>1</sup> 230°C/2160 g<sup>2</sup> Secant @ 1% strain

manufacturer are given in Table 4 and Table 5 as follows:

**Table 5: FORTILENE 1986 Polypropylene Data**

Property	ASTM Test	Typical Values	
		English Units	SI
Units	Method		
Density	D 1505	—	
.900 g/cc			
Melt Flow Index <sup>1</sup>	D 1238	—	50
g/10 min			
Tensile Strength			
@ Yield (2 in/min)	D 638	4700 psi	
32.4 MPa			
@ Break (2 in/min)	D 638	2850 psi	
19.7 MPa			
Elongation			
@ Yield (2 in/min)	D 638	10%	
10%			
@ Break (2 in/min)	D 638	200%	
200%			
Flexural Modulus <sup>2</sup>	D 790	230,000 psi	
1586 MPa			
Notched Izod Impact Strength			
@ 23°C	D 256	0.4 ft-lbf/in	21
J/m			
TUP Impact <sup>3</sup>		48 in-lbf	5.4
J			
(7/1/88)			
<sup>1</sup> 230°C/2160 g			
<sup>2</sup> Secant @ 1% strain			
<sup>3</sup> 1" Radius tip on 1/16" plaque			

The coupling agent was EPOLENE G-3002, a maleated polypropylene wax which was obtained in pellet and powder form from Eastman Chemical Co. of Kingsport, Tennessee. Previous work on coupling agents at the FPL has found the greatest improvement in properties from using G-3002 as compared to other maleated polypropylenes. EPOLENE G-3002 has a higher acid number (reactivity) and higher molecular weight than the other agents studied. The higher molecular weight translates into longer polymer chain lengths, which facilitate greater miscibility and hence better interaction with the polypropylene matrix polymer.

A white titanium dioxide master batch colorant was added to two kenaf/polypropylene blends. The colorant is commercially available from General Color & Chemical Co. of Minerva, Ohio.

The reference drum material was a standard, industrial 55-gallon wound fiber drum commercially available from Sonoco, Inc.

In this Example 1 a total of 8 blends were produced to determine the effects of the following variables on the final composite material of the present invention. These variable are (1) fiber type: (kenaf (K) or jute (J)), (2) coupling agent (0% or 3% by mass of fiber), (3) polypropylene type (Polypropylene-H or Polypropylene-L), and (4) fiber content (40% or 50% by total mass). A tabular representation of these variables is shown in Table 6, Test Design, which provides a quick reference and facilitates comparison of the differences between the blends.

**Table 6: Test Design**

Blend Colorant	Fiber Type	Fiber Content	Coupling Agent	Polypropy- ene Type	
1	K	50%	0%	"H"	
N/A	2	K	50%	3%	"H"
N/A	3	J	50%	0%	"H"
N/A	4	J	50%	3%	"H"
N/A	6	[ SONOCO WOUND FIBER DRUM SAMPLE ]			
N/A	7	K	40%	3%	"H"
N/A	8	K	40%	3%	"L"
N/A	9	K	50%	3%	"H"
2% tot. wt.	10	K	50%	3%	"H"
5% tot. wt.					

The ingredients (fiber, polymer and coupling agent) for each blend were dry-

mixed before being melt-blended. A total of 1.5 kg of material per blend was produced for blends 1-4, 7 and 8. Blends 9 and 10 were 100 g portions of blend 4, where the colorant was added to the blend in the hopper of the injection molder. Blends 5 and 6 were actually sections of the fiber drum cut for testing. Blend 5 was from the drum bottom and blend 6 was from the drum wall. As blend 5 was not representative of the type of material that the composite would be replacing, it was eliminated from the rest of the study.

A 1-L high intensity thermokinetic mixer (K-mixer) was employed for melt-blending the fiber and polymer. For each blend, the batch size was approximately 150 g. Hence, a total of 10 batches were produced for each blend. The average batch time was between 45 and 60 seconds. Dump temperature was set at 200°C, and rotor tip speed was approximately 30 m/s maximum. After melt blending, each batch of a particular blend was granulated and mixed with the other batches of that blend.

The resultant blends were injection molded into standard ASTM test specimens using a Cincinnati-Milacron 33-ton reciprocating screw injection molder. Materials were injection molded with a flat temperature profile of 188°C (370°F), and injection pressures between 1000 and 1300 psi maximum. Once molded, specimens and remaining blend materials were stored over desiccant in sealed bags at 90°F and 20% relative humidity for 3 days before testing.

A portion of the blends were also used to produce curved cylindrical panels for a modified compression test as described earlier. To produce the curved panels for the compression test, 360 g of the granulated material were placed in a mold in a hot press and pressed at 193°C (380°F) at a pressure of 500 psi for 5 min., then at 1000 psi for 1 min.. The final dimensions of the pressed panel were 12" x 12" x

0.138". These panels were sawed down to 10" x 10" square pieces for further forming. The panels were then formed to give a uniaxial curvature equal to that of the fiber drum (27.5 cm radius of curvature). This was accomplished by placing a single panel on a stainless steel form of equal curvature, placing a mated form on the top of the panel, and loading approximately 8 kg of mass on the top form to assist in correct forming. The entire apparatus was then placed in a forced air oven at 150°C for 10 minutes, after which time the curved panel was removed, then cooled and stored under the same conditions as the injection molded test specimens.

The following ASTM standard tests were performed on the composite blends and the drum material:

- Izod Impact Energy (ASTM D256-90b)
  - notched and unnotched (J/m)
- Cantilever Bending (ASTM D747)
  - 9° secant modulus (Gpa)
  - maximum strength (MPa)
- Flexural (ASTM D790-84a)
  - modulus of elasticity (GPa)
  - maximum strength (MPa)
- Tensile (ASTM D638-84)
  - modulus of elasticity (GPa)
  - maximum strength (MPa)
- Ambient Water Soak (ASTM D570-88)
  - dimensional change
  - weight change
  - cantilever modulus and strength

- Melt Flow Index (ASTM D1238-85)

-- samples #2,4,7,8 only.

Five replicate specimens from each blend were used for each test, and average and standard deviations determined. In the ambient water soak test, 1 set of specimens was measured after immersion in water for 24 hours, and a replicate set after immersion for 3 weeks.

The compression tests on the composite material of the present invention and drum material formed was a direct relative comparison between the composite and the drum material. This comparison was made by forming a curved rectangular panel of composite material from a flat, square, pressed composite panel. This panel had the same dimensions (thickness, radius of curvature, length and width) as that of a section cut from the side of the fiber drum. Two pieces of material were placed with convex sides facing each other for stability. The sections were then compressed to failure and the resulting stress versus compression data were measured to obtain maximum axial load.

Finally, the melt flow index was measured using a Kayness melt- indexer. Melt temperature was 190°C and the cylinder load was 2160 g. Standard ASTM conditions for temperature call for 230°C. However, this is above the degradation temperature of the cellulosic fibers (approximately 200°C), and 190°C is standard for similar types of thermoplastic materials. The amount of material extruded out of the cylinder in 10 minutes was measured. The average of 3 trials per sample was determined for blends 2, 4, 7, and 8.

The average results of the mechanical tests for impact, cantilever, flexural, and tensile tests are given in Table 6. Typical curves of the tensile stress-strain relationships for blends 1-4, and 6-8, are given in Figure 3, where the blends appear

at the ends of the stress-strain curves by number. Figures 4-11 compare graphically the mechanical property values for the different tests of the blends 1-4, and 6-8.

Figure 4 compares notched impact energy for each blend of drum materials and composite blend discussed above, while Figure 5 compares the un-notched impact energy for each blend of drum materials and the composite blend. Figure 6 compares the cantilever modulus of elasticity for each drum material and composite blends. Figure 7 compares the maximum cantilever strength for each drum material and composite blend. Figure 8 compares the flexural modulus of elasticity for each drum material and composite blend. Figure 9 compares the maximum flexural strength for each drum material and composite blend. Figure 10 compares the tensile modulus of elasticity for each drum material and composite blend. Figure 11 compares the maximum tensile strength for each drum material and composite blend.

Table 8 lists the results of a statistical analysis based on the student's t-test at the 95% confidence level. The table is used to determine if the mechanical property values of two different blends are statistically different from each other. The confidence level in effect determines the degree of rigor with which the comparison is made. A higher confidence level requires two values to have a higher degree of difference from each other in order to not be considered as equivalent values within the margin of error of measurement. In Table 7, the numbers represent the result of a mathematical operation on the property values as part of the t-test. A number highlighted in bold italics indicates that the two property values that this t-test value represents are significantly different (statistically) from one another. Whether or not a t-test value is deemed significant or not depends on the test design, confidence level, and the error associated with each measurement. To be deemed significant, a test result t-value must be greater than a theoretical t-value based on the experimental



design. In this experiment,  $t$  (theoretical) = 2.306. Values of  $t$  (experimental) greater than this are significant; less than this, not significant. This is the limit of information obtainable from this particular test. This test does, however, indicate which variables are having a true effect on mechanical properties and not simply appearing to have an effect because of experimental errors due to small sample numbers.

Tables 7 and 8 results are as follows:

Table 7. Mechanical Test Results

Blend	Fiber	%	Coupling Agent	Polypropylene	Irod Impact		Cantilever		Flexural		Tensile	
					Notched (J/m)	Unnotched (J/m)	Modulus (GPa)	Max Strength (MPa)	Modulus (GPa)	Max Strength (MPa)	Modulus (GPa)	Max Strength (MPa)
1	Kend	50%	0%	F1986 (50 WFI)	23.67 [1.56]	54.74 [2.40]	3.3 [0.70]	60.32 [1.20]	7.18 [0.33]	53.98 [0.34]	8.3 [0.83]	33.7 [0.83]
2	Kend	50%	3%	F1986 (50 WFI)	26.91 [0.86]	110.03 [8.86]	4.24 [0.57]	95.7 [0.99]	7.08 [0.50]	88.06 [0.87]	8.18 [0.17]	58.12 [0.63]
3	Jule	50%	0%	F1986 (50 WFI)	27.28 [0.43]	73.88 [9.02]	4.06 [0.50]	75.15 [1.21]	7.46 [0.22]	70.97 [1.23]	8.69 [0.13]	46.27 [1.44]
4	Jule	50%	3%	F1986 (50 WFI)	27.64 [1.54]	128.91 [10.47]	3.95 [0.08]	98.72 [2.59]	7.23 [0.22]	93.79 [1.16]	8.4 [0.20]	63.65 [0.88]
6	Sunoco Wound Fiber Drum Sample				215.91 [16.22]	256.06 [38.16]	1.55 [0.05]	31.88 [2.46]	2.25 [0.32]	20.24 [1.99]	3.32 [0.08]	27.68 [0.70]
7	Kend	40%	3%	F1986 (50 WFI)	28.52 [0.79]	137.63 [12.75]	2.53 [0.03]	82.03 [0.69]	5.07 [0.25]	75.45 [1.29]	5.94 [0.58]	49.33 [0.70]
8	Kend	40%	3%	F1602 (12 WFI)	28.39 [1.06]	153.13 [7.44]	2.66 [0.05]	88.91 [0.84]	5.07 [0.15]	80.21 [0.67]	5.64 [0.04]	52.57 [0.29]
9	Kend	40%	3%	F1986 (50 WFI)	n/o	n/o	3.58 [0.00]	89.55 [2.42]	6.68 [0.16]	81.38 [0.85]	n/o	n/a
10	Kend	50%	3%	F1986 (50 WFI)	n/o	n/o	3.40 [0.06]	84.82 [8.74]	6.24 [0.96]	80.89 [0.96]	n/o	n/a

\*Note: Numbers in brackets represent the standard deviations of the measured values based on 5 sample replicates.

Table 8. Results of Statistical Analysis

Values Compared		1 and 2	3 and 4	7 and 4	1 and 3	7 and 8	2 and 7
Significance		Effect of G-3002 on Kenaf Material	Effect of G-3002 on Jute Material	Effect of Jute vs. Kenaf (with G-3002)	Effect of Jute vs. Kenaf (without G-3002)	Effect of MFI on Kenaf Material	Effect of Fiber Content (40% vs. 50%)
Mechanical Properties							
Notched Impact		4.07	7.50	0.93	1.92	0.22	3.08
Unnotched Impact		13.47	8.90	3.08	4.59	3.11	3.25
Conf. Secant Modulus		2.33	0.49	1.13	1.98	4.99	6.70
Conf. Max. Strength		50.85	18.44	2.44	19.46	14.15	25.33
Flexural Modulus		0.37	1.65	0.61	1.58	0.00	8.04
Flexural Max. Strength		81.58	30.18	8.84	29.77	7.32	18.12
Tensile Modulus		0.32	2.72	1.87	1.04	1.15	8.29
Tensile Max. Strength		52.40	23.03	11.42	16.91	9.56	20.87
24-Hour Soak Testing							
% Weight Change		11.93	5.39	3.14	7.3	n/a	n/a
% Length Change		1.08	0.96	2.07	2.3	n/a	n/a
% Thickness Change		4.33	2.46	2.69	4.72	n/a	n/a
% Width Change		1.26	1.14	1.21	1.06	n/a	n/a
3-Week Soak Tests							
% Weight Change		40.85	11.48	20.07	21.57	3.25	31.00
% Length Change		3.91	8.03	2.62	2.48	0.33	2.50
% Thickness Change		6.60	5.18	3.97	8.27	4.67	5.08
% Width Change		4.94	0.99	0.40	6.74	3.99	1.02
Notes:							
1. Numbers in the table represent the experimental t-values from comparison of blends listed.							
2. Numbers in bold italics represent a significant difference in property value for the two compared blends.							
3. 1-theoretical - 2.305							

Referring again to Figure 3, an examination bears out several interesting and important fundamental differences in the various composites and drum material. These points are discussed in more detail below, but in brief, the following trends are evident. Composites of jute fibers have greater max. strength and % elongation at break than do composites of kenaf fibers (1 & 2 vs. 2 & 4). The effect of coupling agent is to increase the maximum strength of both types of composites, but have negligible effect on Young's modulus or % elongation at break (1 vs. 3 or 2 vs. 4). Composites with lower fiber loading (7, 8 & 9 vs. 2 & 4), generally have lower modulus and maximum strength but greater % elongation at break, except when compared to 50% kenaf filled composites, in which % elongation is greater.

Based on the above results in Tables 7 and 8 and Figures 3-10, the following points are noteworthy. Jute fiber typically performs better than kenaf fiber as a reinforcement in polypropylene (compare blends 1 vs. 3 or 2 vs. 4). This is particularly evident when examining the maximum strengths of these materials. There is less of a difference in the modulus between composites of these two fibers. This is supported by the statistical analysis of table 3. These results follow behavior of individual kenaf and jute fibers, with the jute being stronger than kenaf, but both of relatively similar modulus. The effect of jute vs. kenaf fibers on impact energy is negligible.

Comparisons of blends 1 vs. 2 and 3 vs. 4 shows the effect of adding EPOLENE G-3002 coupling agent to the fiber/polypropylene composite at a level of 3% by weight of fiber. In either comparison, the results clearly show the benefit of adding coupling agent on the maximum strength property of the composites. On the other hand, the modulus is relatively unaffected by the presence of coupling agent. The statistical analysis supports these observations. These results

are as expected, since at very low strains where the modulus is measured, the stiffness depends mainly on the stiffness of the fiber, and less on the stress transfer between matrix and fiber. At significantly larger strains, the maximum strength depends, in a large part, on the efficiency of transferring the stress from the relatively weak matrix to the much stronger fiber. The coupling agent acts to increase the bonding and stress transfer between matrix and fiber, and thus will have little effect on modulus but a significantly greater effect on the maximum strength of the composites. The effect of coupling agent on impact energies was to slightly increase the notched impact energy (which is a measure of crack propagation), and more significantly increase the unnotched impact energy (which is a measure of crack initiation), as compared to blends without coupling agent.

The effect of using a high melt flow index, high stiffness polypropylene homopolymer (Polypropylene-H) as the matrix polymer compared to the low melt flow index homopolymer (Polypropylene-L) is shown in the comparison of blends 7 and 8. Notched impact energy values are statistically the same, while the lower melt flow index polymer system (blend 8) shows approximately a 20% increase in unnotched impact energy over blend 7. In cantilever, flexural, and tensile maximum strength tests, the lower melt flow index system performed better, statistically, than did the high melt flow index system. However, the high melt flow index system showed a statistically significant increase in Young's modulus over the low melt flow index system of approximately 5%. The flexural modulus and cantilever modulus were not significantly different. These results can be explained in part by examining the properties of the matrix polymers.

The composites in this Example 1 tend to follow the same trends illustrated in

manufacturer reported properties of Fortilene 1602 (F1602-(Polypropylene-L) - 12 melt flow index polymer and) Fortilene 1986 (F1986-(Polypropylene-H) - 50 melt flow index polymer) as presented in Tables 4 and 5 - namely, little disparity in modulus between the two polymers. The reason for the smaller amount of disparity in modulus of the composites than would be expected based on matrix polymer considerations alone, can probably be explained by the fact that cellulose itself can act as a nucleation site for crystallinity. If this is the case, the high nucleation of densely filled composite systems such as these would negate the theoretical advantage of the more crystalline polymer. Hence, the final properties tend not to be as different from one another as would be expected by considering matrix polymer effects alone.

The above circumstance is also illustrated in the results of the melt flow index tests. Table 9 summarizes the results. For practical purposes, blends 2, 4, and 8 (50% kenaf/, 50% jute/, and 40% kenaf/12 melt flow index polypropylene, respectively), are quite similar. Blend 7 (40 % kenaf/50 melt flow index polypropylene) has a melt flow index 1.8 times that of blend 8, while the corresponding unfilled polypropylene has a nominal (190°C) melt flow index over 4 times that of F1906 (used in blend 8). These composites have melt flow index values between 1 and 2.5, which is considerably less than the polymers alone. This is due to both the lower temperature (190°C) of the test and the fact that the effect of high fiber loading is predominant over that of the melt flow index of the matrix. To effectively enhance processability, other processing aids, such as lubricants, may be helpful.

**Table 9: Melt Flow Indices for Selected Composite Blends**

Blend	Fiber	% Fiber	Coupling Agent	Polypropylene	MFI gms/10 min
2	Kenaf	50%	3%	F-1986 50 MFI	1.35 [0.07]
4	Jute	50%	3%	F-1986 50 MFI	1.53 [0.01]
7	Kenaf	40%	3%	F-1986 50 MFI	2.48 [0.07]
8	Kenaf	40%	3%	F-1602 12 MFI	1.37 [0.13]

\* Note: Numbers in brackets represent the standard deviations of the measured values based on 5 sample replicates

Blends 2 and 7 are a direct comparison of the effect of fiber content (by mass) on properties. Both contain kenaf fiber with 3% G3002 with blend 2 at 50% by mass of fiber and blend 7 at 40% by mass of fiber. As would be expected for a less filled system, blend 7 shows higher values of impact energy. Composite failures is due to the initiation and propagation of cracks in the material. Fibers have regions of high stress concentration near their ends and tend to promote crack growth, which leads to a lower energy failure. It is easier for a crack to propagate along a fiber-matrix interface and in regions of high stress than it is through a plastic matrix. Also to be expected, blend 2 shows significantly higher values of strength and modulus for all three types of mechanical tests of this type. As mentioned previously, strength and stiffness are partly dependant on the properties of the reinforcing fiber. Larger amounts of fiber would lead to higher values of strength and stiffness for a well coupled system.

Blends 9 (2% colorant) and 10 (5% colorant) can be compared to blend 2 to obtain a rough estimate of the effect of adding a colorant to the composite blend. The data in Table 2 show that adding the colorant produces a significant drop in modulus and strength of about 10% on average. A greater decrease is indicated as percentage of colorant increases to 5%. The appearance of such colored composites was that of a slightly brownish gray opaque tint with some surface fiber showing, with a smooth and glossy surface appearance. The 5% colored material had a whiter appearance than did the 2% material.

The results of the ambient water soak test are presented in Table 10 and graphically in Figures 12 - 17. Listed are the blends and their corresponding percent changes (from dry to wet) in weight, thickness, length and width of standard flexural bar specimens. Also shown are the changes in cantilever beam maximum strength and secant modulus. The percent changes in this case were calculated by comparing wet values to values obtained in the mechanical tests from the first part of the study. The 24 hour water soak test included only blends 1-4 because there was not enough of the original blends 7 and 8 to do both a 24 hour and 3 week test. The 3 week test was chosen preferentially over the 24 hour test for blends 7 & 8 to emphasize long term effects. The fiber drum samples were not subjected to the soak test since they lost their integrity within 24 hours.



Table 10. Ambient Water Soak Test Results

Percent Changes in Dimensional and Mechanical Properties After 24 Hours Soaking											
Blend	Dimensional Stability (% Change)				Conilveer Secant Modulus (GPa)			Conilveer Max. Strength (MPa)			
	Mass	Length	Width	Thickness	Before	After	% Delta	Before	After	% Delta	
1	1.86	0.012	0.35	1.89	3.3	2.82	-14.55	60.32	54.13	-10.26	
2	1.20	-0.030	0.00	0.72	4.24	3.31	-21.93	95.7	89.63	-7.39	
3	1.37	0.076	0.10	0.065	4.06	3.42	-15.76	75.15	69.18	-7.94	
4	1.08	0.050	0.22	0.26	3.95	3.64	-7.85	98.72	90.52	-9.20	
Percent Changes in Dimensional and Mechanical Properties After 3 Weeks Soaking											
Blend	Dimensional Stability (% Change)				Conilveer Secant Modulus (GPa)			Conilveer Max. Strength (MPa)			
	Mass	Length	Width	Thickness	Before	After	% Delta	Before	After	% Delta	
1	9.83	0.201	2.91	7.37	3.3	1.7	-48.48	60.32	43.81	-27.37	
2	5.43	0.133	1.26	4.37	4.24	2.25	-46.93	95.7	68.22	-28.71	
3	5.86	0.249	1.37	4.36	4.06	2.33	-42.61	75.15	60.54	-19.44	
4	3.95	0.084	1.12	2.71	3.95	2.79	-29.37	98.72	81.03	-17.92	
7	2.46	0.074	0.82	1.59	2.53	2.23	-11.86	82.03	73.44	-10.47	
8	2.11	0.066	0.59	0.96	2.66	2.46	-7.52	88.91	80.62	-9.32	

The data indicate that after 24 hours, and especially after 3 weeks, of soaking in ambient water, there are differences between jute and kenaf and in blends with and without coupling agent. Overall dimensional changes are very low, typically below 5%, and without coupling agent, less than this. The values for changes in thickness and width are of an order of magnitude greater than that of the length. This is most likely due to the fact that fibers were generally aligned along the length of the test samples, and that width and thickness are dimensions in which fibers swell when absorbing moisture.

All the data except for percent change in length without coupling agent, show that jute fibers outperform kenaf fibers in giving the smallest percentage change in properties after soaking.

Most significant is the effect of coupling agent on the percentage change in properties. EPOLENE G-3002 significantly decreases the change in dimensions and mass, along with significantly decreasing the losses in modulus and strength. Coupling agents tend to promote better bonding between fiber and matrix, and thus inhibit water molecule penetration and migration along the fiber-matrix interface. The compression test results are listed in Table 11.

**Table 10: Compression Test Results**

Blend	Fiber	% Fiber	Coupling Agent	Polypropylene	Avg. Load to Failure
2	Kenaf	50%	3%	F-1986 50 MFI	1,200 kg. [160]
4	Jute	50%	3%	F-1986 50 MFI	1,200 kg. [170]
Sonoco Wound Fiber Drum Sample				880 kg. 50 MFI	[50]
<p>* Note: Numbers in brackets represent standard deviations of the measured values based on 3 replicates</p>					

As can be seen in Table 11, both the jute and the kenaf composites performed in a similar manner. The jute and kenaf composites show an approximately 40% increase in maximum axial load over the fiber drum. This indicates that this material should be able to perform at least as well as a similarly designed wound fiber drum in terms of ability to support axial loading.

One of the most significant results is the comparison of the mechanical properties of the new jute/ or kenaf/polypropylene composite materials with those of a typical wound fiber drum material. In this particular instance, the thickness of the supplied fiber drum material was found to fall within the specified range of thickness for the molded plastic parts to which they were compared ( $3.2 \pm 0.4$  mm). In an extreme case, the fiber drum thicknesses were greater than the average thickness of the composite specimens by only a few percent. This is acceptable for experimental conditions in this study, since if the composite can perform better than a thicker drum section, then it should also perform better than a section of the same thickness.

The data shown in Table 11 lead to a number of conclusions. The fiber drum material

performed significantly better than either the jute or kenaf composite material in the Izod impact test. Although the Izod impact test standard used in this study was for plastic materials, this would tend to be a good indication of the relative performance of the drum material compared to the composite material. Polypropylene and fiber composites of the nature of those in this Example 1 tend to be somewhat more brittle than would be expected of the fibrous material composing the Sonoco drums. The impact properties of these composites are by no means optimized due to the method of compounding used in Example 1. The impact performance can be further enhanced to yield results exceeding the results for the fiber drum material by adding impact modifiers or increasing the fiber lengths of the fiber reinforcements by improved processing methods. A few of the impact modifiers may include the addition of various rubber compounds and EVA, to name a few.

The modulus of the composite material is increased over that of the drum material by over 150% for the cantilever and tensile tests, and to over 200% for the flexural test. The maximum strengths of the composite material is increased over that for the drum material by 130% for the tensile test to over 360% for the flexural test. The results for the modulus and maximum strength correlate with the results of the compression test, since the inward failure of the proposed drum of the specified length to 20 radius ratio is dictated by stiffness more so than by buckling of the drum under axial load.

Conclusion suggested by the example are as follows: Composites of Jute/ or kenaf/virgin polypropylene homopolymer can be produced at levels of at least 50% fiber with typical plastics molding equipment. Composites of this nature show significant improvements over wound fiber drum material in the areas of modulus, maximum strength, and maximum axial load in

compression. Improvements of these properties range from 40% for axial compression load to up to 150%-350% for modulus and maximum strength in composites of jute with coupling agent. Composites of this nature perform at levels significantly lower than that of the fiber drum material when subjected to Izod impact tests. The drum material shows improvement over the composite material from 100% to over 360% in unnotched impact energy versus the range of composite blends at 50% fiber to over 650% in notched impact energy versus the range of composite blends at 50% fiber. However, a direct comparison of impact performance using the Izod impact test may not be warranted with respect to the nature of final use of these materials in the end product considered for production.

Jute fibers prepared according to the present invention act as better reinforcing fibers as compared to kenaf fibers in polypropylene composites examined in this study, especially when considering the maximum strength of the composites. When subjected to an ambient water soak test, jute fiber composites also show smaller dimensional and mass changes, as well as smaller decreases in cantilever modulus and maximum strength, than do composites made with kenaf fibers.

The effect of using a coupling agent in these composites is to significantly increase the maximum strengths and impact properties, while the moduli are relatively unaffected. Addition of coupling agent also significantly decreases the changes in dimension and mass, as well as showing decreases in the loss of cantilever maximum strength and modulus as compared to composite systems without coupling agent. There was little significant overall effect of using a 50 melt flow index nucleated, high modulus polypropylene homopolymer matrix compared to using a non-nucleated 12 melt flow index polypropylene homopolymer in terms of mechanical properties

and process ability. In this case the effect of high fiber loading outweighs the effects of polypropylene crystallinity variations.

Using 50% fiber loading by mass compared to 40% loading of kenaf/polypropylene composites with coupling agent gave the expected results of lower impact energies but higher values of modulus and strength for the 50/50 composite blend. The addition of 2% or 5% by total mass of white titanium dioxide master batch colorant in 50% kenaf composites produced a blend with acceptable surface and color qualities. The mechanical properties of such composites were somewhat decreased by addition of colorant. For most industrial applications, colorant can be omitted, and the containers can be painted where color and identifying legends and instructions are required.

Referring to Figure 18, a generalized chart illustrating the relationship between fiber loading and composite strength is shown. The relationship between fiber length/mean critical fiber length and efficiency is illustrated in Figure 18. About 90% of theoretical strength is achieved at 1.5 times the mean critical fiber length - after which very little improvement occurs. This is probably in the neighborhood of an aspect ratio of 100 - or 5 mm fiber length. Generally, pullout length is inversely proportional to the shear-bond strength. The critical length/diameter ratio is twice the pullout length/diameter ratio.

Other calculations include the bond Shear Strength which equals the tensile strength/4 times the length/diameter ratio. The Pullout length/diameter ratio is equal to the tensile strength/4 divided by Bond Shear Strength. The critical length/diameter ratio is simply two times the pullout length/diameter ratio. For jute-like fibers a critical length/diameter ratio of from about 50 to about 150 is a good range, depending on the strength of bond achieved by the coupling agent.

In general, minimal loading levels should be achieved, particularly for short fibers. Otherwise they can only act as filler material. There is preferably an interaction between fibers. This interaction only begins to occur when there is significant overlap between the areas of reduced stress surrounding individual fibers.

At levels of addition of fiber above 45% to 50% as a percentage of fiber by weight, the addition of more fiber will generally reduce the composite strength due to packing, distribution and wetting problems.

The addition of filler material at low loading levels will have the effect of shifting the curve upwards, because of an increase in composite tensile modulus due to the addition of filler. This provides better stress transfer across fibers than does the matrix itself.

Injected jute-like fibers have a specific gravity close to unity, which is significantly lower than that of other competitive synthetic reinforcing material. The reinforcing value of jute-like fibers per unit weight is significantly higher than that of glass fiber - 2.5 specific gravity vs 1.29 for jute and a higher tensile strength and tensile modulus. A specific gravity of filler which is close to the specific gravity of the plastic material does not add much weight by volume, as opposed to glass, which will add significantly to weight by volume. Jute-like fibers also hold strength-for-weight advantages over other materials.

Table 12 compares the tensile, flexural, and impact properties, along with water absorption and specific gravities, of the neat polypropylene, the 50%-kenaf composite, and several commonly utilized injection molded filled/reinforced polypropylene materials. The comparison is meant as a general indicator and not as an exact comparison, which would be possible only if all the specimens were prepared and tested under identical conditions.

The tensile and flexural properties of the kenaf-filled polypropylene show significant improvement over those of the unfilled polypropylene. The tensile modulus of the kenaf-fiber system is approximately equivalent to that of the 40%-mica polypropylene or to a 30% chemically coupled glass-fiber polypropylene composite (from the data on 30%- and 40%-glass- filled polypropylene), but significantly higher than that of the talc and calcium carbonate-filled compounds.

**Table 11: Properties of Polypropylene Composites**

Filler/reinforcement							
Property	None	Kenaf	Talc	CaCO <sub>3</sub>	Glass*	Glass*	Mica
% filler by weight	0	50	40	40	30	40	40
% filler by volume	0	40	18	18	8	19	18
Tensile modulus, GPa	1.7	7.2	4	3.5	5	9	7.6
Tensile strength, MPa	33	62	35	25	83	110	39
Elongation at break, %	>>10	2.2	--	--	2.5	2.5	2.3
Flexural strength, MPa	>>41	88	63	48	97	131	62
Notched Izod impact, J/m	24	32	32	32	98	107	27
Specific gravity	0.9	1.07	1.27	1.25	1.05	1.23	1.2
Water absorption, %/24 hr.	0.02	0.95	0.02	0.02	0.02	0.06	0.0

\* Chemically coupled glass fibers.

With respect to the data of Table 12, the kenaf filaments were obtained from Agro-Fibers, Inc., and cut into lengths of about 120 cm. The fibers, polypropylene (Solvay 1602), and a maleated polypropylene (Eastman Chemical Co. G3002) were blended in a thermokinetic mixer, granulated, dried, and then injection molded into standard ASTM specimens. The blend contained 50% kenaf fibers, 1.5% maleated polypropylene, and 48.5% polypropylene (weight percent). Data



for talc and  $\text{CaCO}_3$  from Modern Plastics Encyclopedia, 607 (1989). Data for glass and mica from "Materials Selector 1993," Maeter. Eng., 174 (1992).

A composite made of polypropylene and kenaf fibers extracted from a commercial crop has mechanical properties comparable with those of commercial polypropylene composites. The low density of the kenaf-polypropylene composite offers materials cost savings, but the high water absorption of the composite means that care is required in the selection of applications.

As was seen in Table 12, a Comsat composite made of polypropylene and kenaf fibers extracted from a commercial crop has mechanical properties comparable with those of commercial polypropylene composites.

Flow under convergent conditions, under the correct circumstances, can lead to axial unidirectional orientation, and flow under expanding condition leads to orientation perpendicular to the flow. Mold design can, where possible incorporate these concepts to achieve certain fiber orientation. Fiber orientation is achieved at the injection nozzle and can be maintained throughout the flow under conditions of injection from thick to thinner part. Mold design can be optimized to achieve this intended effect. In one instance, to pick one of many, in a die containing ribbing, the channels used to create the ribbing can be used to concentrate and channel injected material to other parts of the die. This produces fiber alignment and greater strength.

Random fiber orientation is enforced through the addition of micro spheres [explain micro spheres, what they are and how to add them] that provide enhanced fiber distribution - essentially better micro-packing. This has the effect of providing better stress transfer from fiber to fiber. The fiber resists strain locally, inducing a much higher stress in the fiber than in the surrounding matrix.

The use of fibers having an aspect ratio of 200 and which are oriented, improve tensile stress loading performance by up to 500% over orthogonal fibers. Transverse (orthogonal) orientation of the fibers results in failure at between 0.6% (557 aspect ratio) and 1.0% (185 aspect ratio) of the strain, and at between 1/8th (557 aspect ratio with 1.6% strain at failure) and 1/6th (185 aspect ratio with 1.4% strain at failure) of the stress applied to oriented fiber composite.

Water absorption is a function of the bond, coating and crystallinity of the polypropylene. The more amorphous the polypropylene, the less water absorption will occur. Note that with high degree of orientation and high fiber loading the more ductile, amorphous polypropylene composite behaves almost identically to the more crystalline polypropylenes. With a more ductile matrix, transverse alignment will still result in a small increase in tensile strength such that, in a brittle matrix, it will cause a decrease in tensile strength with fiber loadings. This is probably because of stress concentration in the transverse direction.

Jute or kenaf/polypropylene composites can even better serve in the materials for drum containers in the food shipping industries, with further modifications. The impact performance of the composite of the present invention could be increased by the addition of elastomeric impact modifiers in the polypropylene matrix material. This method could prove cost prohibitive on a commercial basis, however. Another means to improving the impact properties of this type of composite is to increase the minimum fiber lengths by appropriate processing conditions while maintaining adequate dispersion. This could be accomplished by the use of less intensive mixers such as continuous kneaders or appropriately configured twin screw extruders.

Other techniques which hold promise include the evaluation of the use of recycled materials as raw materials for composite manufacture. Even considering the composite of the

present invention alone, it may be recycled with itself in amounts up to 50%. Thus, once materials are placed into service, any materials which become damaged in service may be returned to the manufacturing site for recycle blending into freshly made materials.

Other cost effective coupling agents in composite manufacture can be used. While EPOLENE G-3002 has heretofore shown to produce the greatest improvements in properties, there may be other more inexpensive coupling agents beyond the scope of this work that may offer adequate property enhancements. More controlled blending and molding can also help to control the marginal properties of the final composite product. Larger machines with a slower blend revolution speed can help preserve fiber length. While the present invention has been described in terms of a composite and method for making the composite, one skilled in the art will realize that the structure and techniques of the present invention can be applied to many materials and processes. The present invention may be utilized in any situation where additional strength and other advantageous characteristics are sought from forming a composite from natural fibers and a plastic material.

Although the invention has been derived with reference to particular illustrative embodiments thereof, many changes and modifications of the invention may become apparent to those skilled in the art without departing from the spirit and scope of the invention. Therefore, included within the patent warranted hereon are all such changes and modifications as may reasonably and properly be included within the scope of this contribution to the art.

WHAT IS CLAIMED:

1. A composite structure comprising:

from about 20% to about 92% by weight of a thermoplastic which has been treated with at least one compound selected from the group consisting of:

maleic anhydride; and

acrylic anhydride; and

from about 8% to about 80% by weight of at least one fibrous substance selected from the group consisting of:

jute;

kenaf;

roselle;

aramina; and

sunnhemp (*crotalaria*)

and wherein said fibrous substance is dispersed throughout said thermoplastic.

2. The structure recited in claim 1 wherein said length of said fibers are from about 1 mm to about 20 cm.

3. The structure recited in claim 1 wherein said thermoplastic is at least one thermoplastic selected from the group consisting of polypropylene and polyethylene.

4. The structure recited in claim 1 wherein said fibers contain lignocellulosic fiber and

wherein at least some of said lignocellulosic fiber is acetylated.

5. The structure recited in claim 1 wherein said composite structure exists in at least one of an injection-molded, blow molded, press molded, vacuum molded, and rotation molded state.

6. The structure recited in claim 1 and further comprising a wall made from said composite structure.

7. A method of manufacturing a composite material comprising the steps of:  
contacting (57) a natural fiber (51) with a maleated thermoplastic to form a fiber/ maleated thermoplastic blend or preform; and  
blending (59) an additional amount of an unmelted thermoplastic with the fiber/maleated thermoplastic preform to form said composite material.

8. The method recited in claim 7 wherein an average length of said natural fibers (51) is from about 1 mm to about 20 cm.

9. The method recited in claim 7 wherein said thermoplastic is at least one thermoplastic selected from the group consisting of polypropylene and polyethylene.

10. The method recited in claim 7 wherein said fibers (51) contain lignocellulosic material

and further comprising the step of acetylation of at least some of said lignocellulosic fiber.

11. The method recited in claim 7 wherein said thermoplastic has a melt flow index of from about 8 to about 50.

12. The method recited in claim 7 wherein said fiber/ maleated thermoplastic preform is produced comprising the step of mixing maleic anhydride with said powdered thermoplastic.

13. The method recited in claim 12 wherein said maleic anhydride is present with said powdered thermoplastic in a concentration of from about 3% to about 6% by weight.

14. The method recited in claim 12 and further comprising the step of adding t-butyl peroxyester to catalyze the reaction of said powdered thermoplastic and said maleic anhydride to form maleated polypropylene.

15. The method recited in claim 14 wherein said t-butyl peroxyester is present in a concentration of about 3% by weight.

16. The method recited in claim 12 wherein said reaction of said powdered thermoplastic and said maleic anhydride is conducted for 2 hours in an inert atmosphere at a temperature of from about 120° to about 130°C.

17. The method recited in claim 7 wherein said blending (59) step is performed by mixing the maleated thermoplastic with the natural fiber(51).

18. The method recited in claim 7 wherein said blending (59) step is performed by the free fall attachment of the maleated thermoplastic onto the natural fiber(51).

19. The method recited in claim 18 wherein said free fall attachment is performed while said maleated thermoplastic is electrically charged.

20. The method recited in claim 7 wherein said blending step (59) is performed by dipping said natural fiber (51) into maleated thermoplastic.

21. The method recited in claim 20 wherein dipping step is performed by dipping a continuous bat of fiber (51) through an emulsion bath.

22. The method recited in claim 20 wherein said emulsion bath further comprises:

about 20% to about 40% by weight maleated polypropylene;

about 50% to about 70% by weight water;

about 2% to about 4% by weight potassium hydroxide;

about 0.3% by weight sodium metabisulfite;

and about 5% to about 8% by weight surfactant.

23. The method recited in claim 22 wherein said surfactant is nonylphenol.

24. The method recited in claim 7 and further comprising the step of injection molding (65) said composite material.

25. The method recited in claim 24 wherein said injection molding (65) step is performed at a temperature of about less than 230° C.

26. The method recited in claim 25 wherein said injection molding (65) step is performed at a pressure of about less than 28,000 pounds per square inch.

27. The method recited in claim 24 and further comprising the step of poulticing said composite material before said injection molding (65) step.

28. The method recited in claim 7 wherein said contacting a natural fiber (51) with a maleated thermoplastic and said blending an additional amount of an unmelted thermoplastic is performed simultaneously by dry mixing(57).

29. The method recited in claim 7 wherein said blending an additional amount of an unmelted thermoplastic with the fibrous maleated thermoplastic is performed by adding from about 80% to about 40% by weight of thermoplastic to from about 20% to about 60% fibrous maleated thermoplastic.



30. The method recited in claim 7 and further comprising the step of adding a coupling agent during said contacting a natural fiber with a maleated thermoplastic step.

31. The method recited in claim 9 and further comprising the step of producing maleated polypropylene by mixing from about 3% to about 6% by weight maleic anhydride with from about 97% to about 94% by weight powdered polypropylene.

32. Producing a molded article from a jute-like fiber reinforced thermoplastic composition comprising the steps of:

evenly blending a composition containing between 80% and 99% by weight natural jute-like fibers (51) and between 1% and 20% maleated polypropylene as one of a powder, emulsion and melted polymer;

heat-fixing a bond between the jute-like fibers and maleated polypropylene, to form maleated polypropylene coated fiber;

chopping the maleated polypropylene coated fibers to lengths between 1 and 40 mm;

adding semi-crystalline polypropylene to said maleated polypropylene coated fiber in an amount of from 90% to about 20% of said semi-crystalline polypropylene to from about 10% to about 80% maleated polypropylene coated fiber, to form a composite material; and

injection molding (65) said composite material into a mold to form said molded article.

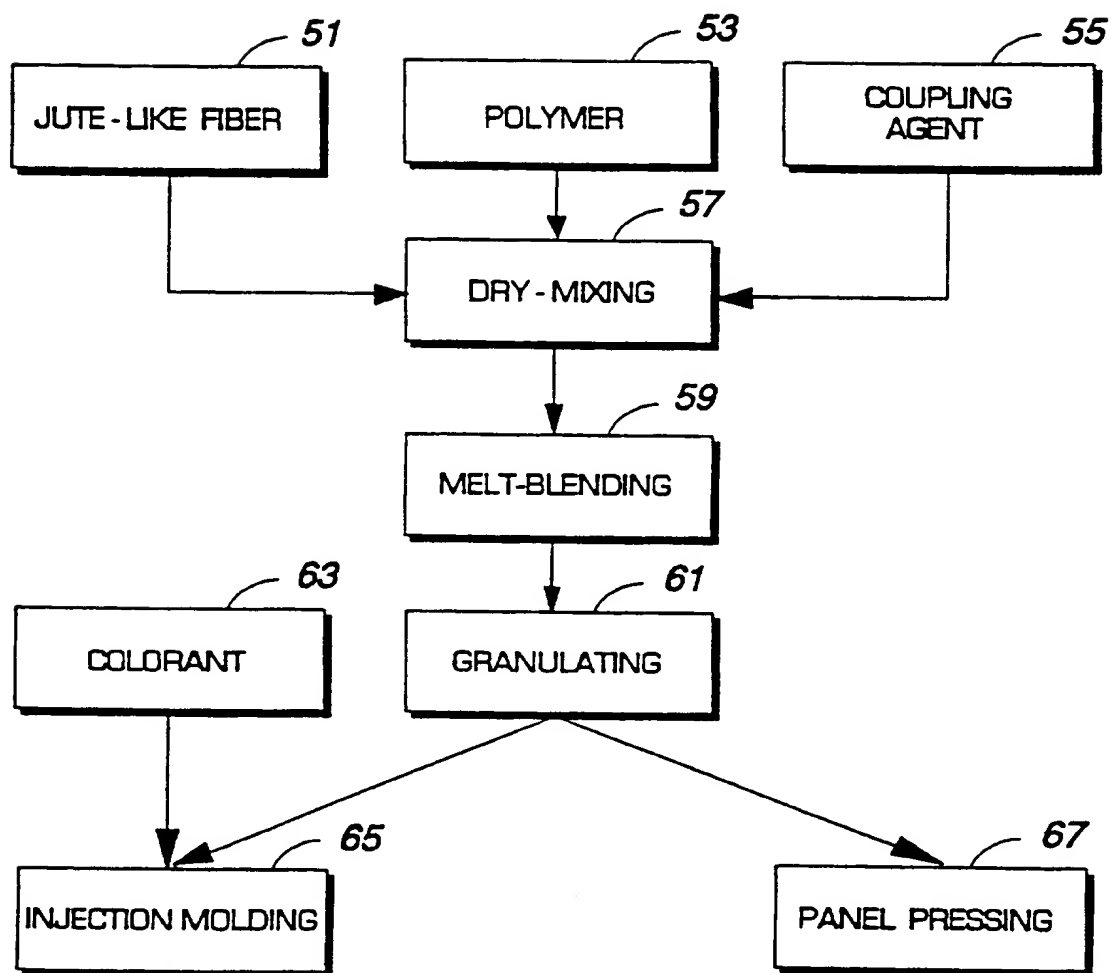
33. The method recited in claim 32 wherein said injection molding(65) step is performed

at a temperature of about less than 230° C.

34. The method recited in claim 32 wherein said injection molding (65) step is performed at a pressure of about less than 28,000 pounds per square inch.

35. The method recited in claim 32 and further comprising the step of pelletizing said composite material before said injection molding step.

36. The method recited in claim 32 wherein said injection molding (65) step may be either press molding, extrusion forming, blow molding, vacuum molding, or blow molding.

*FIG. 1A*

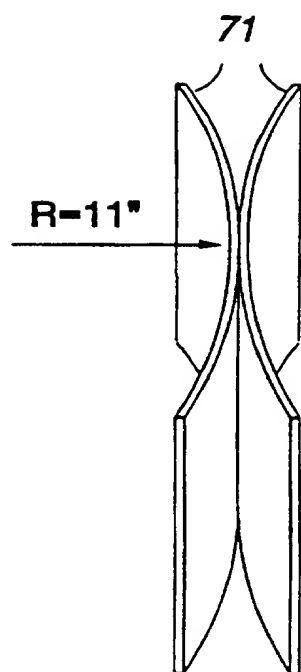
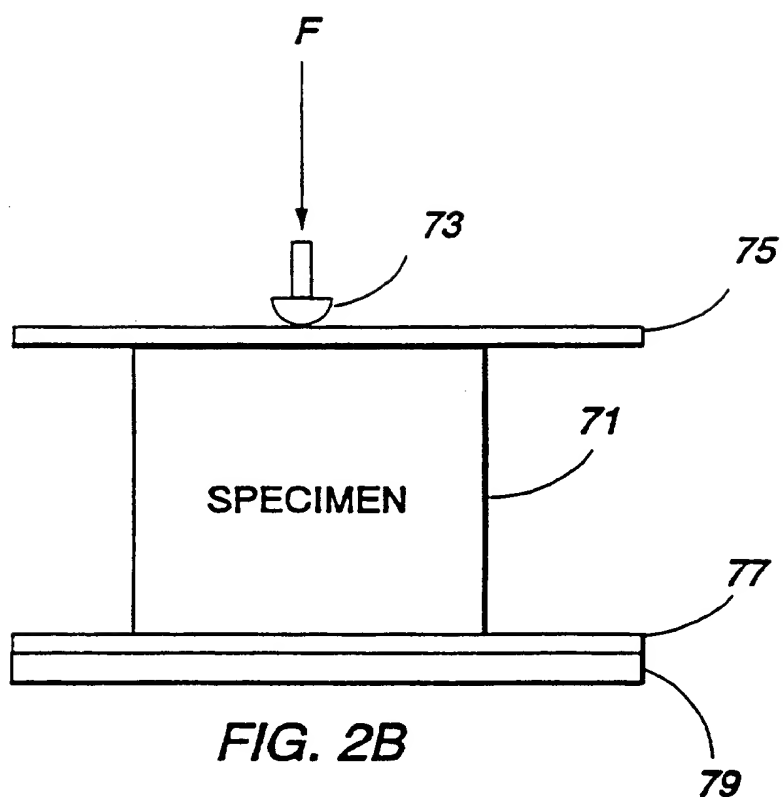
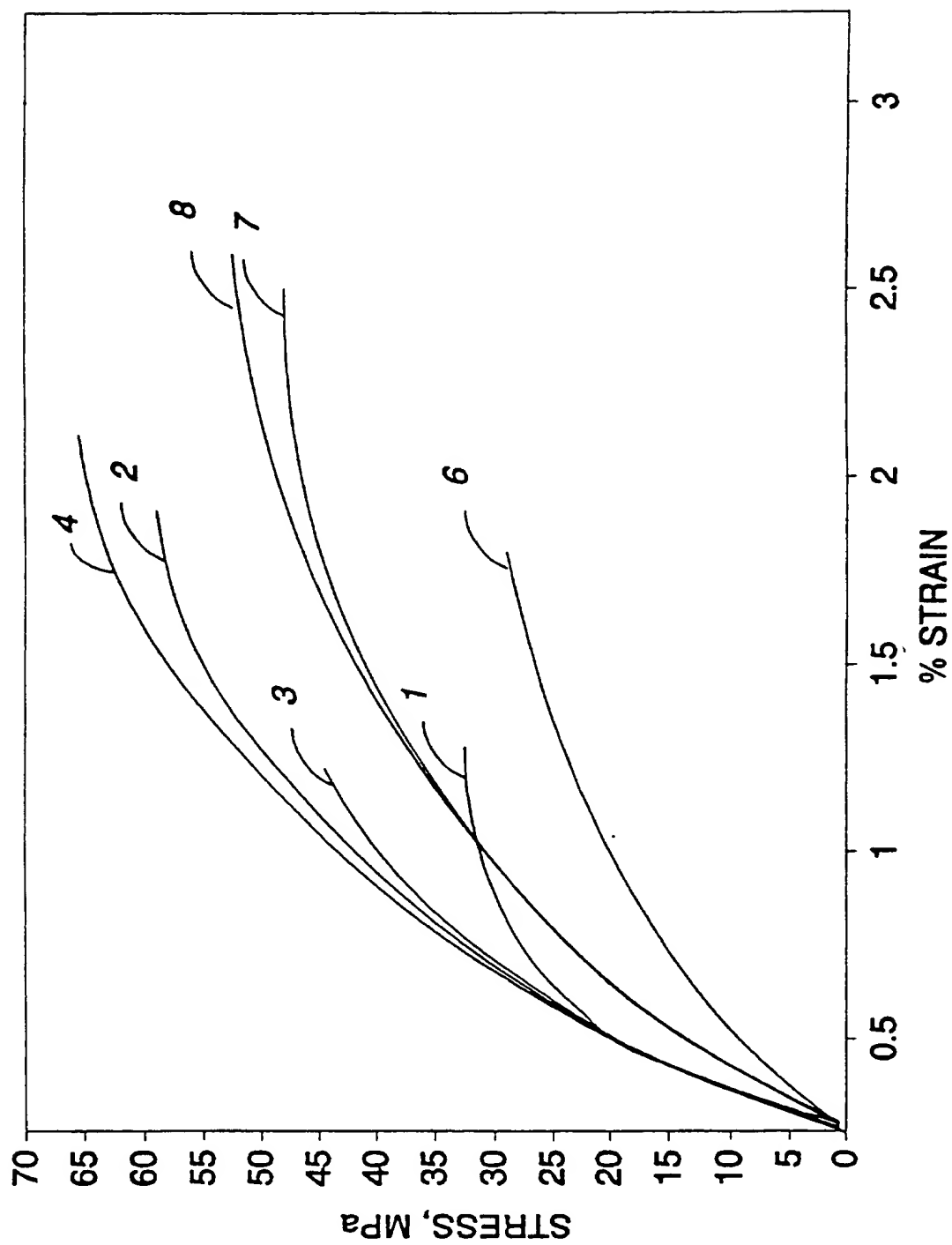
**FIG. 2A**

FIG. 3



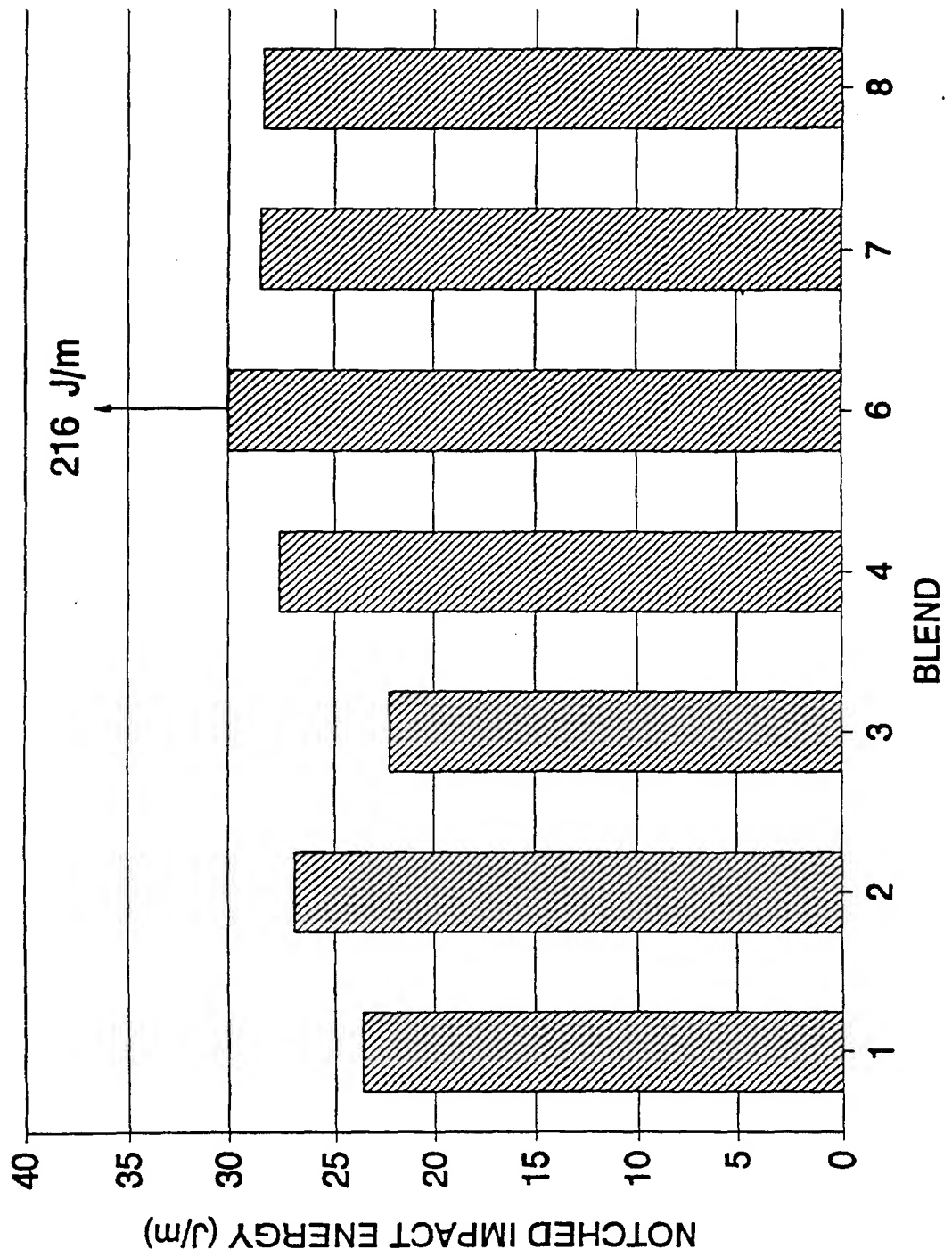
**FIG. 4**

FIG. 5

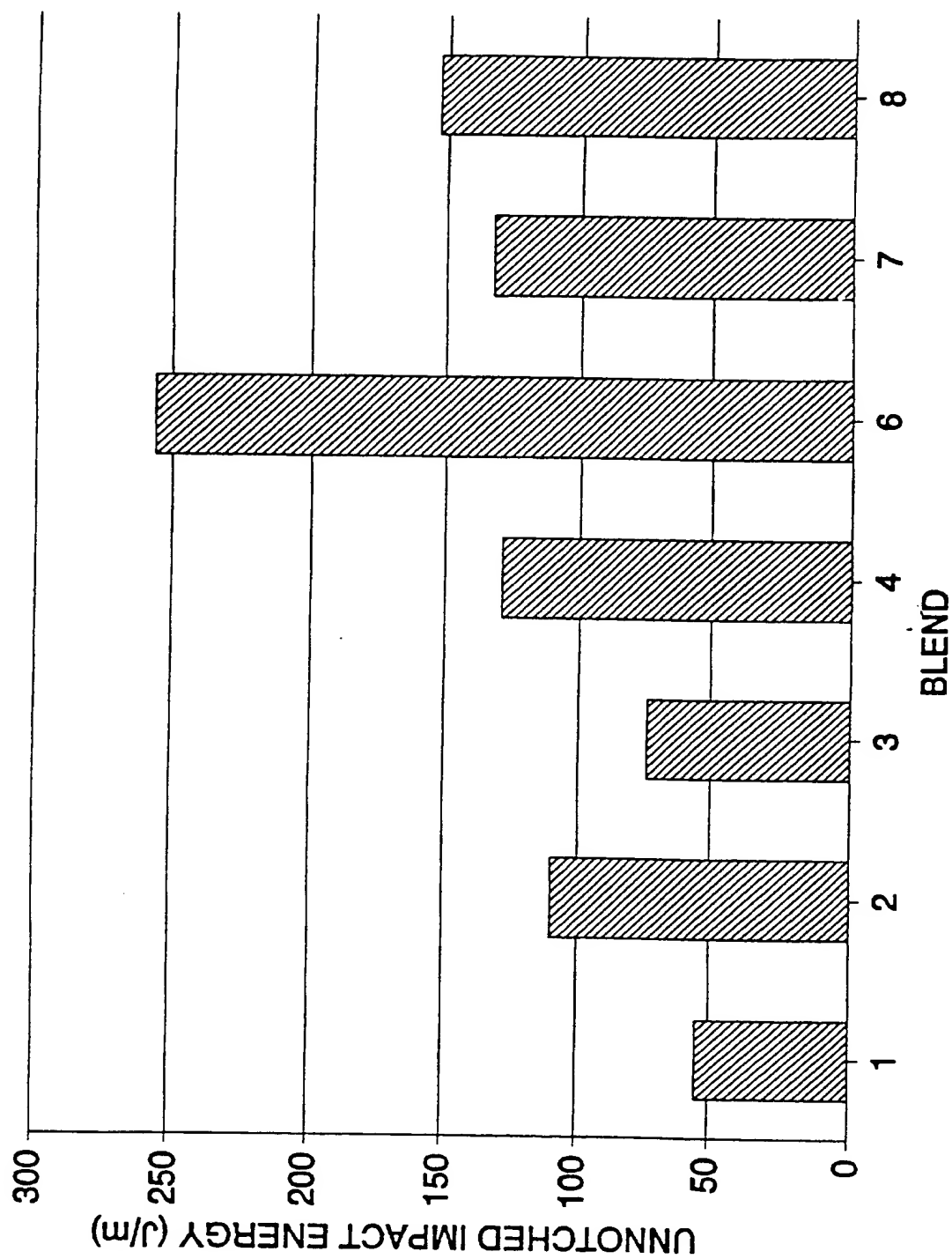


FIG. 6

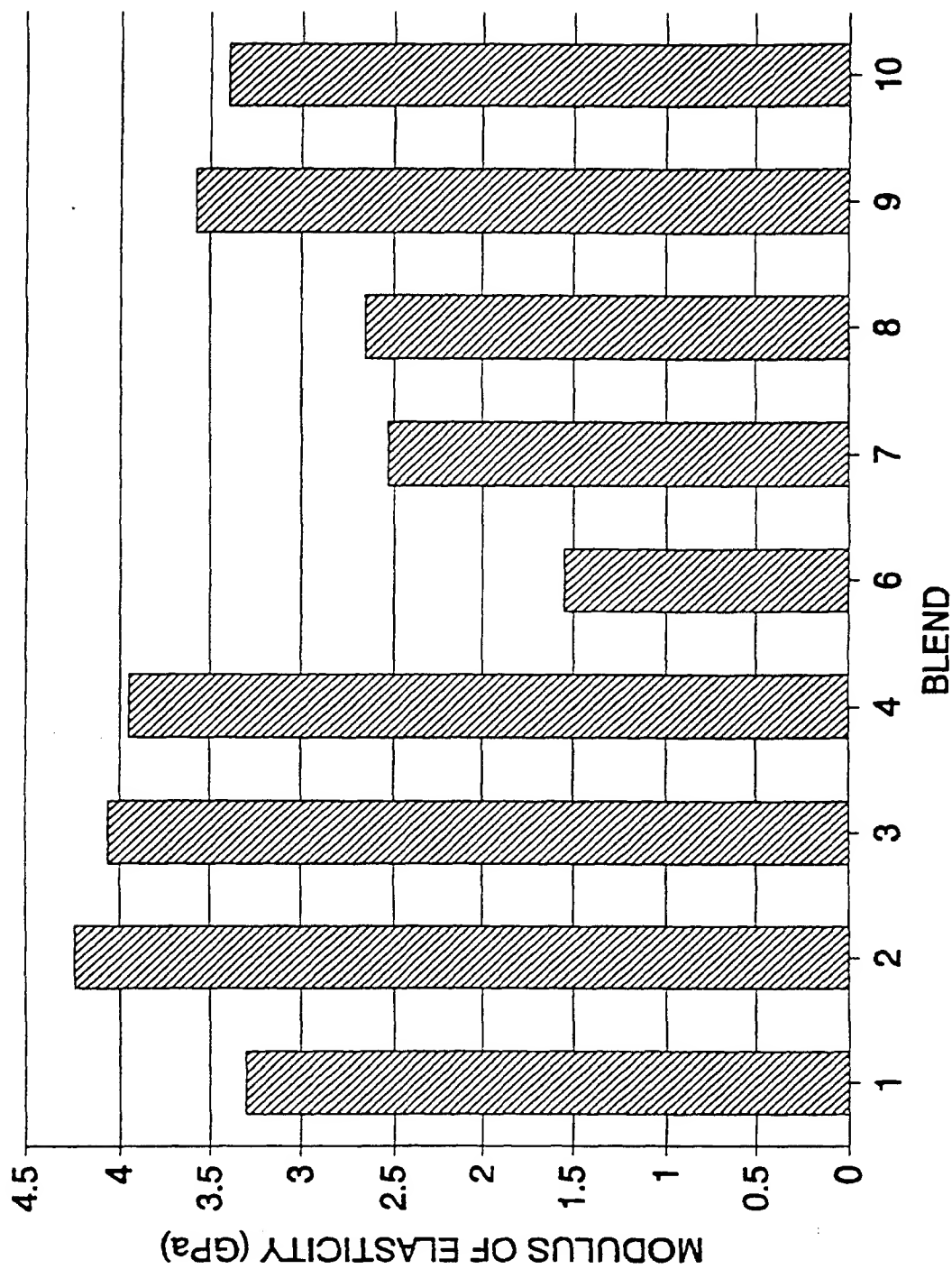




FIG. 8

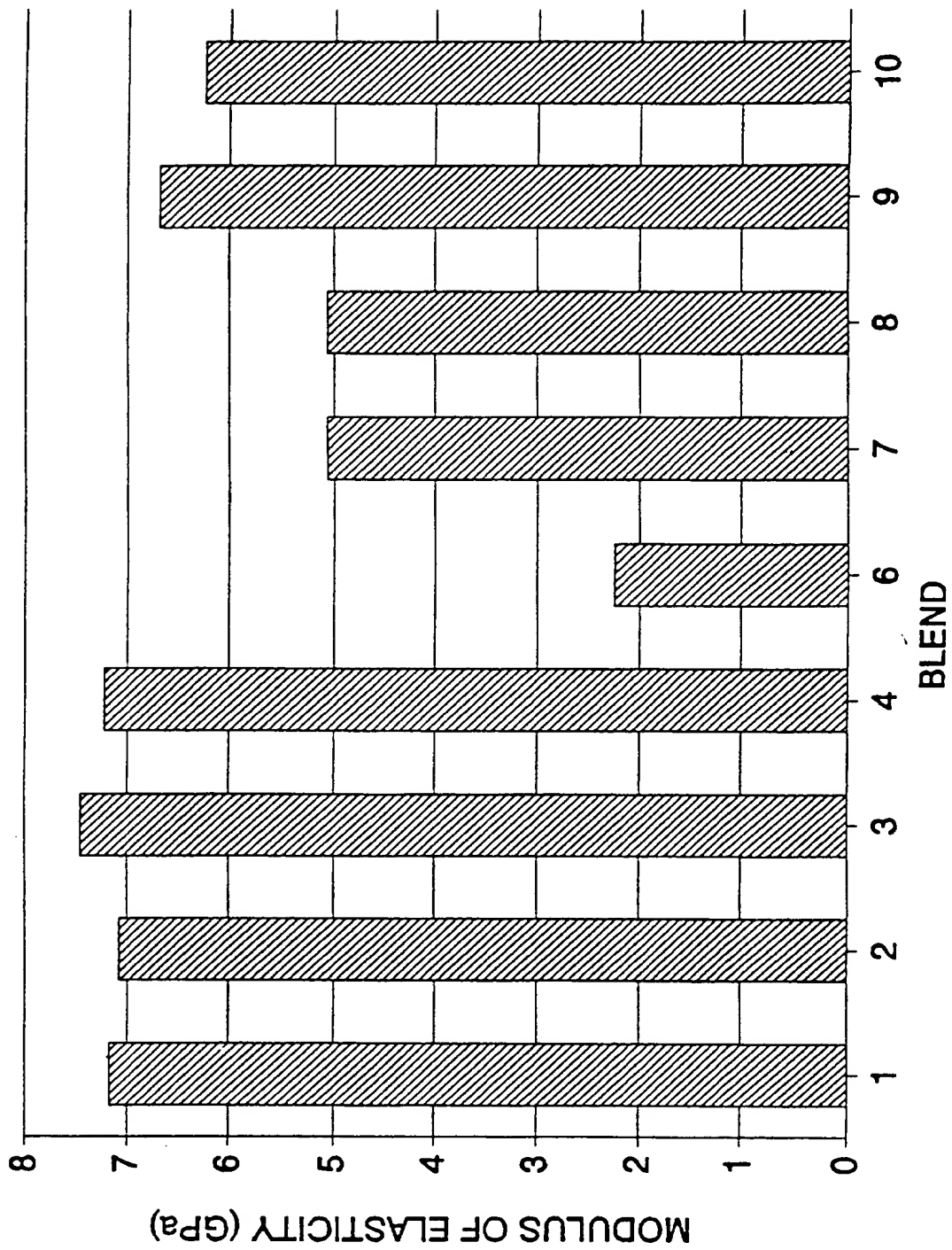
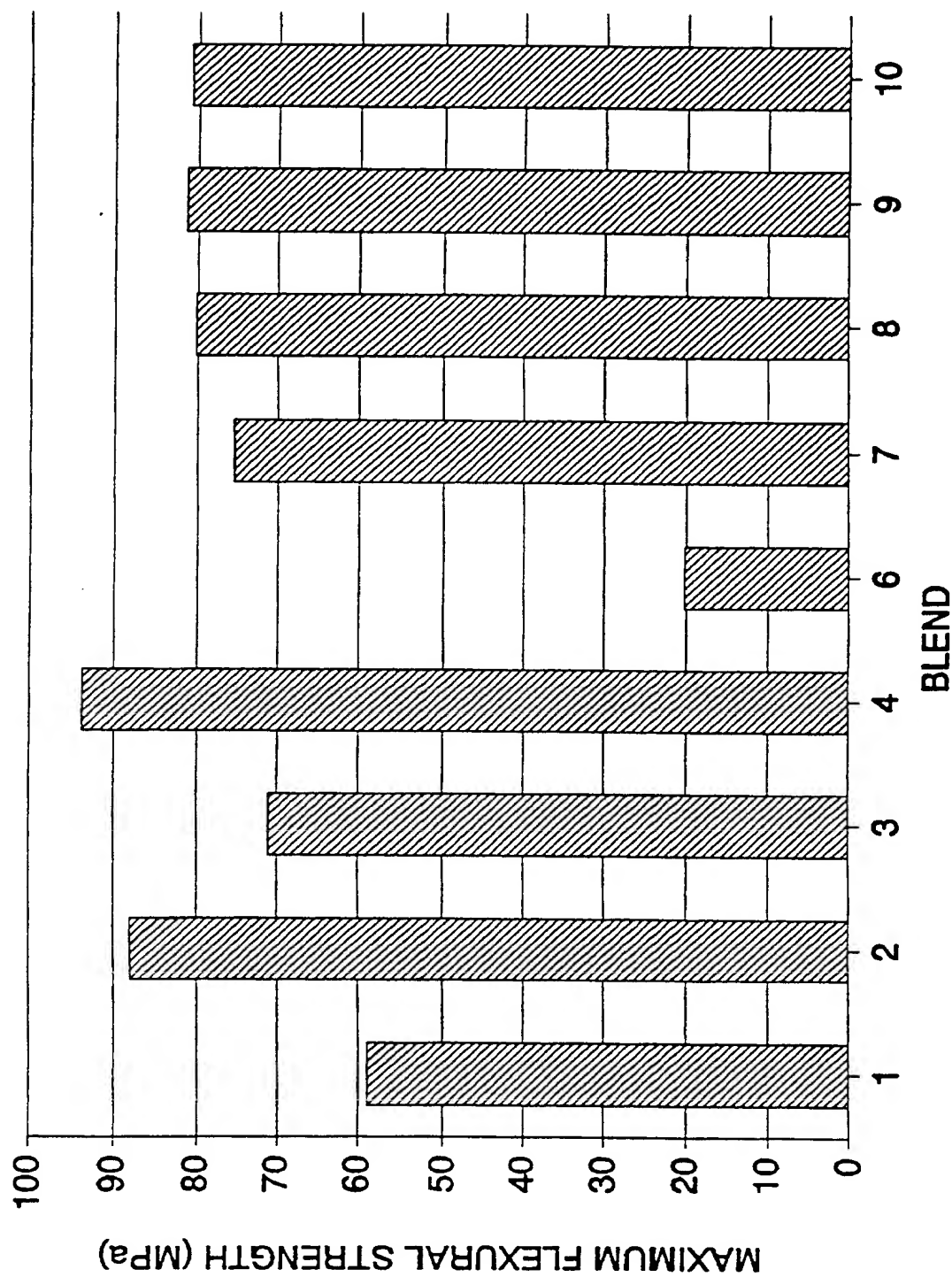


FIG. 9



10/18

FIG. 10

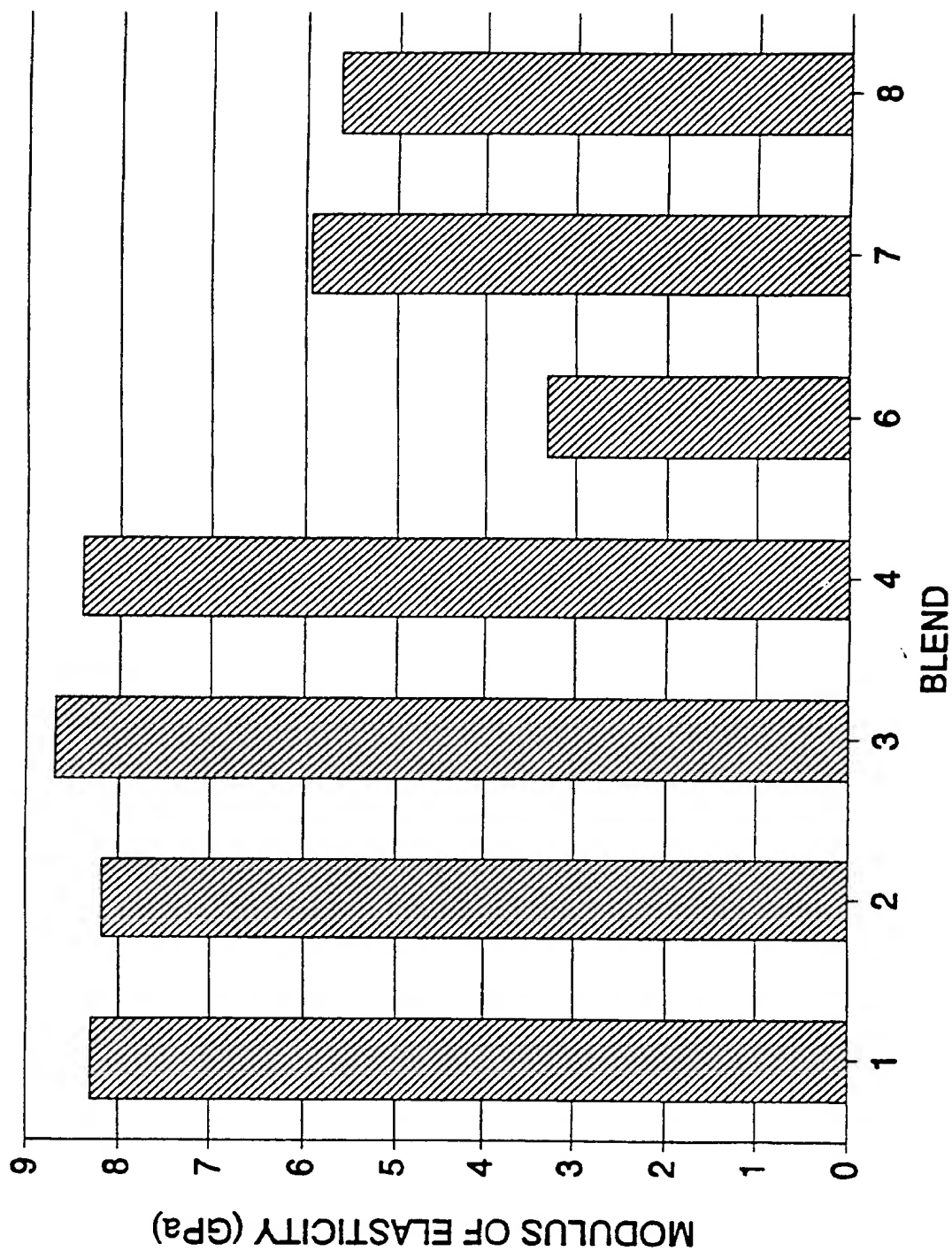


FIG. 11

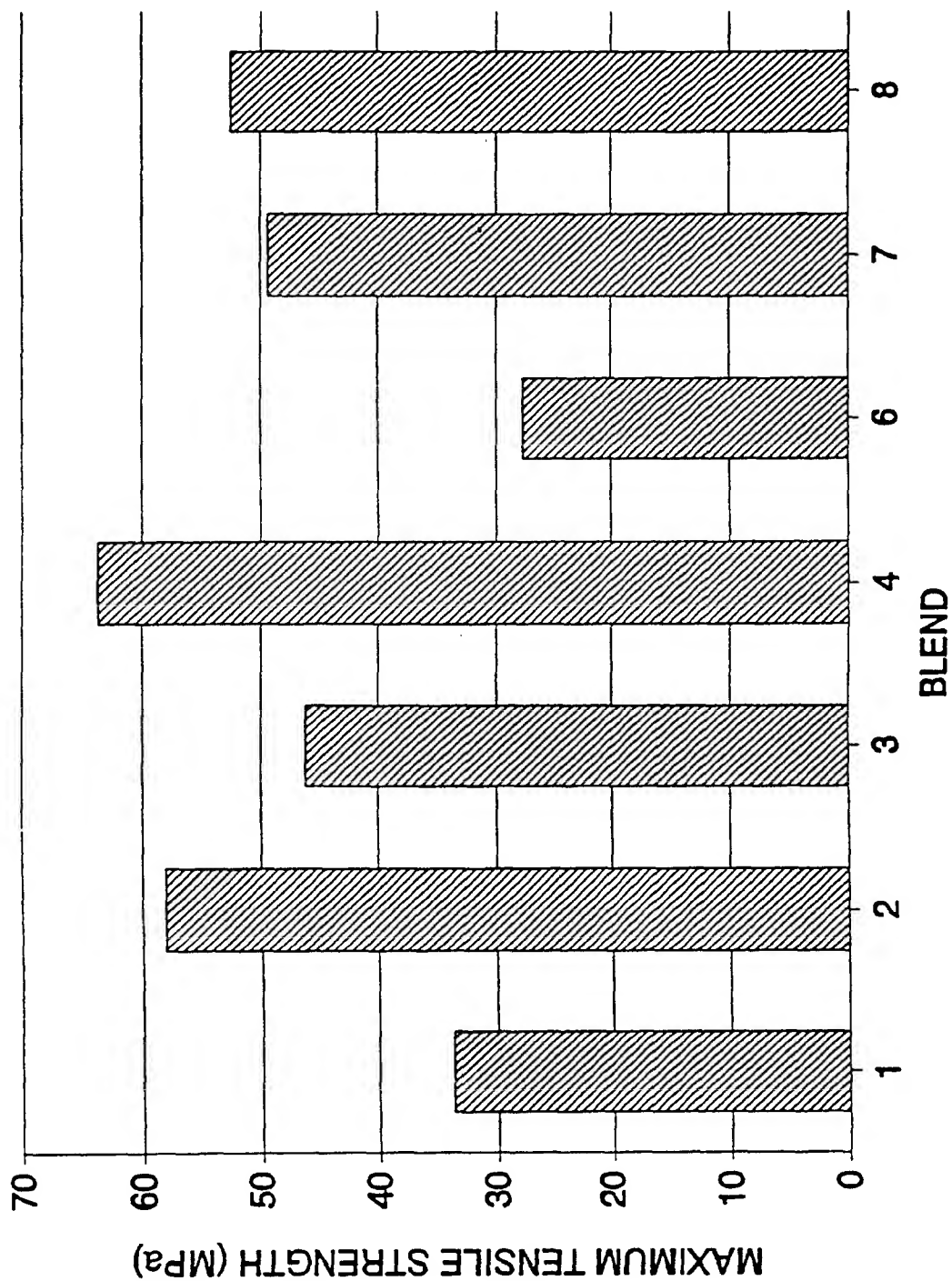


FIG. 12

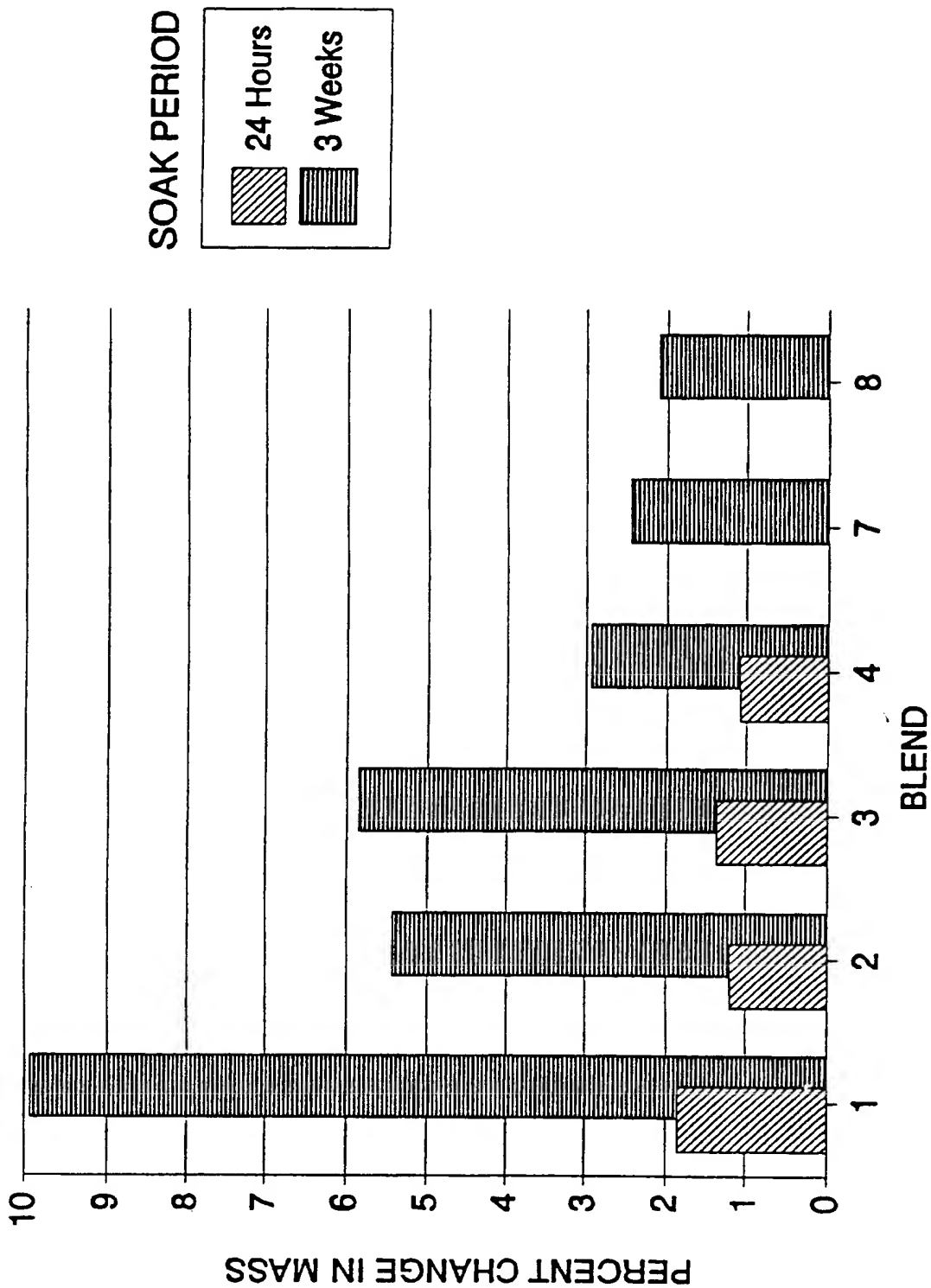


FIG. 13

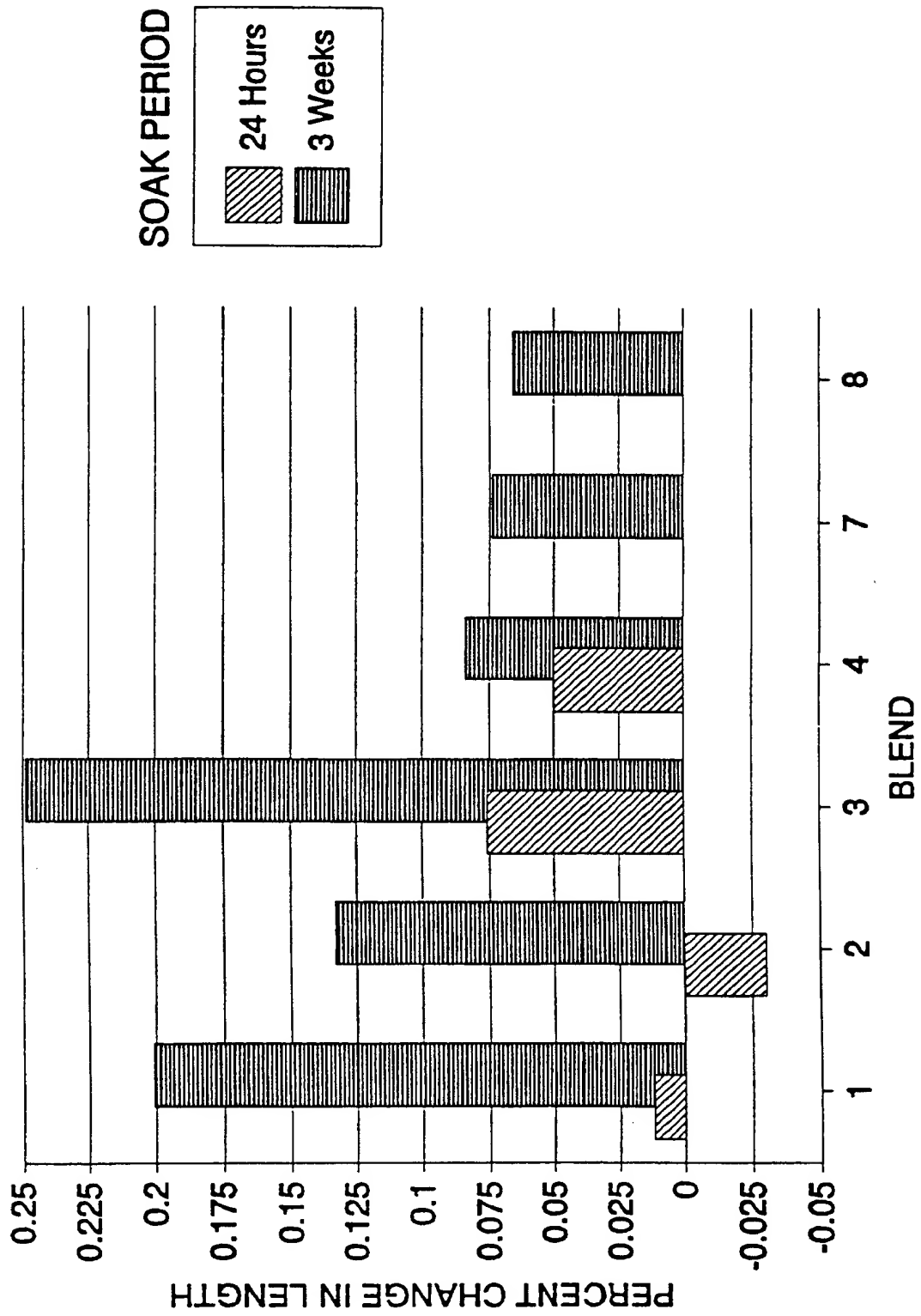


FIG. 14

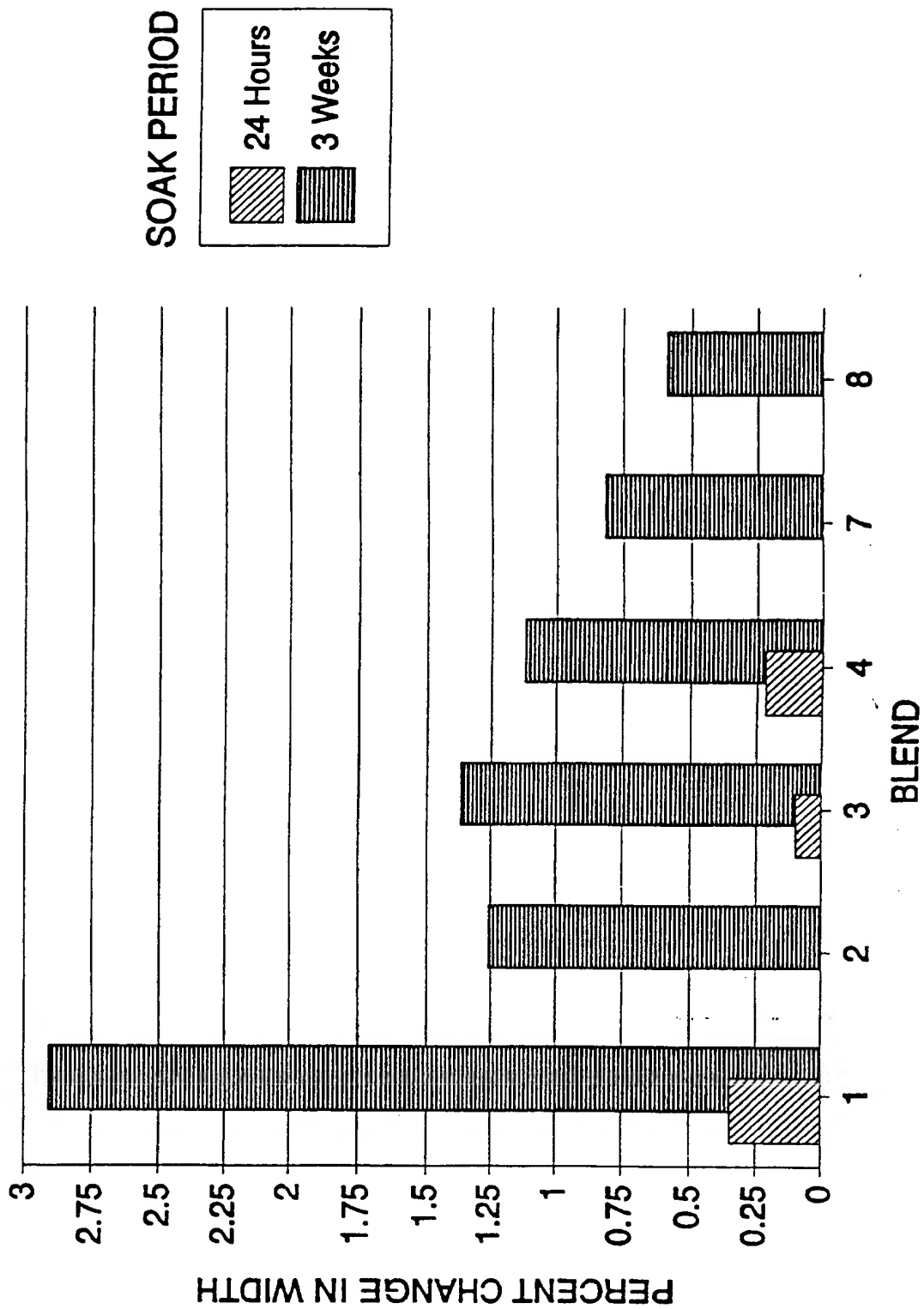


FIG. 15

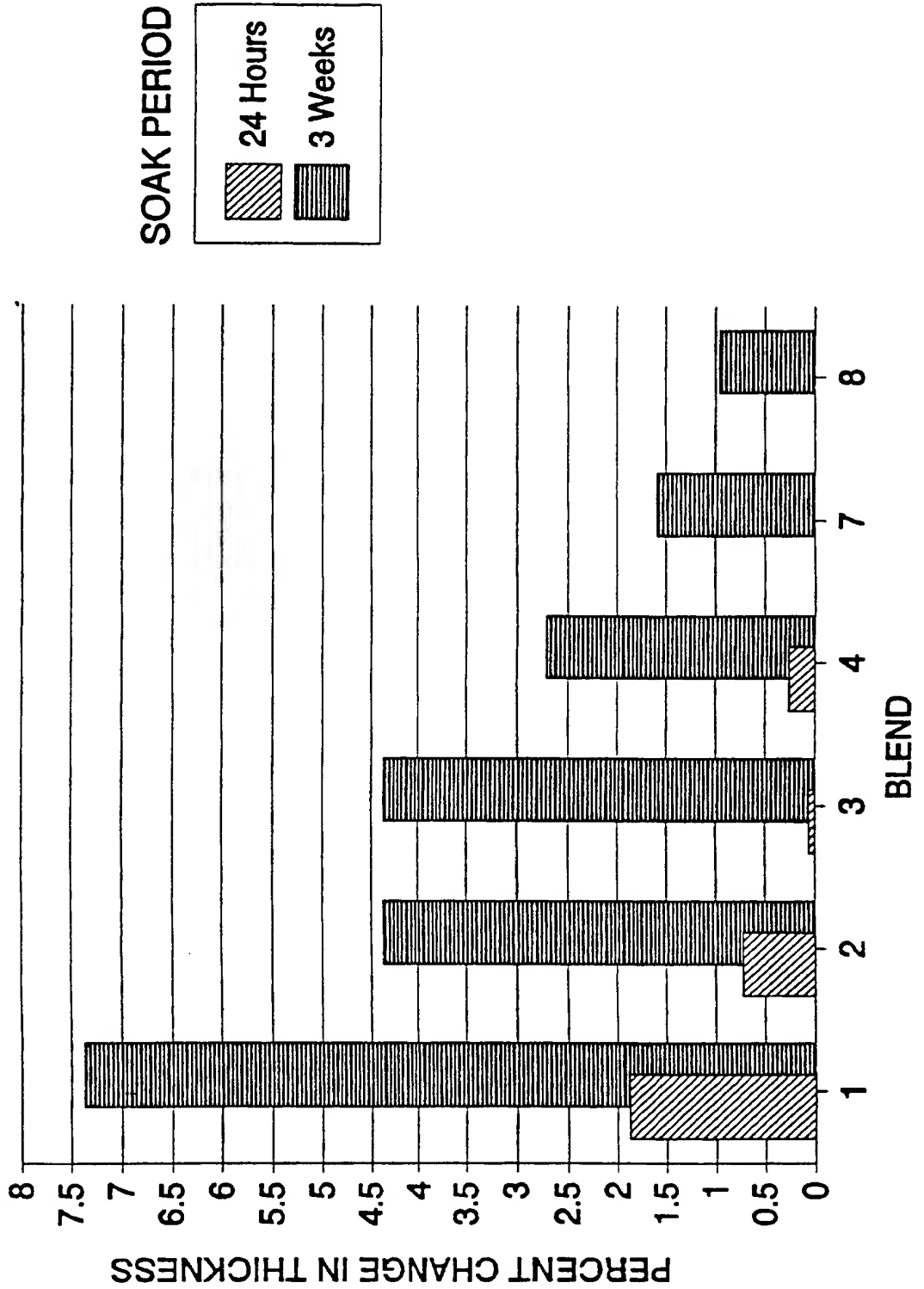




FIG. 16



FIG. 17

